Extending the design space in solvent extraction – From supercritical fluids to pressurized liquids using carbon dioxide, ethanol, ethyl lactate, and water in a wide range of proportions

Veronika Pilařová [a†], Said Al Hamimi [b†], Larissa P. Cunico [b], Lucie Nováková [a], Charlotta Turner [b*]

[a] *Charles University, Department of Analytical Chemistry, Faculty of Pharmacy in Hradec Králové, Heyrovského 1203, CZ-500 05 Hradec Králové, Czech Republic.*
[b] *Lund University, Department of Chemistry, Centre for Analysis and Synthesis, P.O. Box 124, SE-22100 Lund, Sweden.*
† Shared first co-authorship
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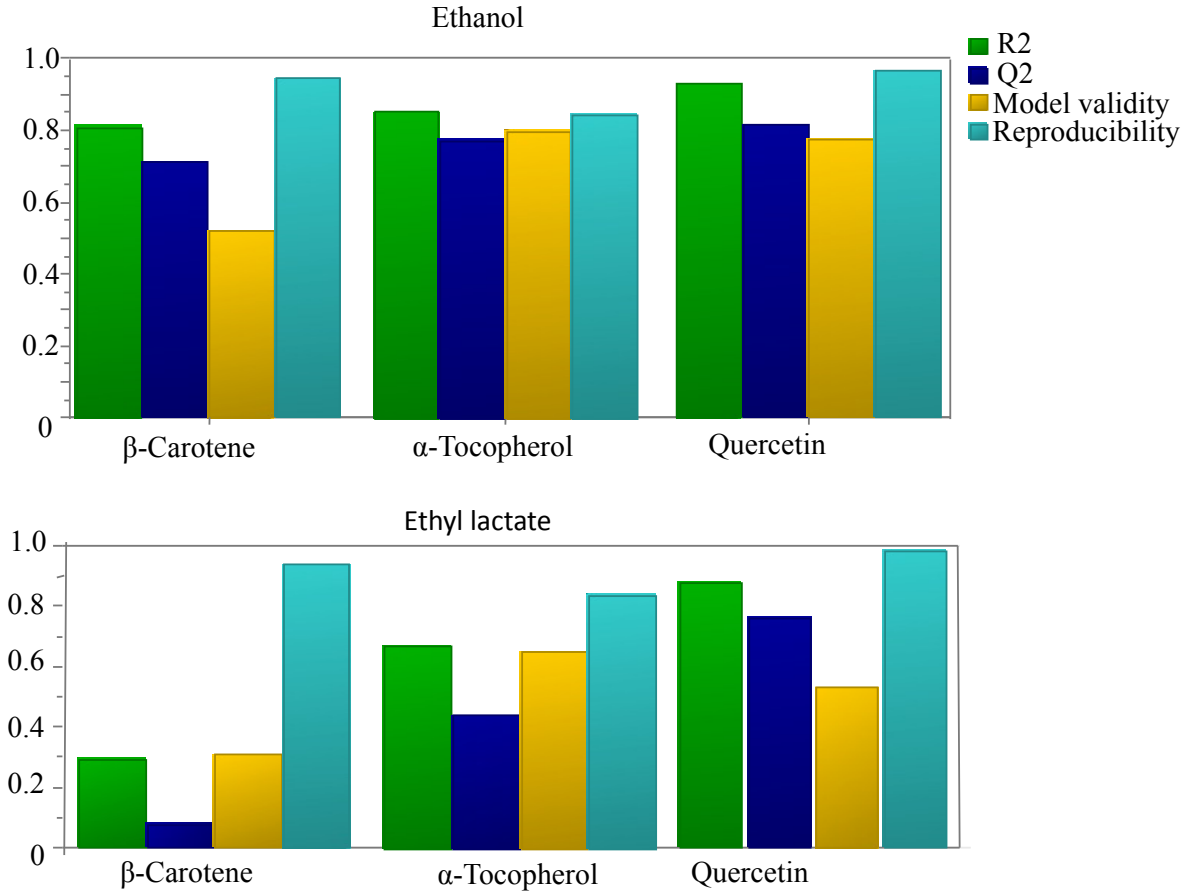

Figure S1. The fitting summary of extraction models for β-carotene, α-tocopherol, and quercetin using ethanol and ethyl lactate as a co-solvent.

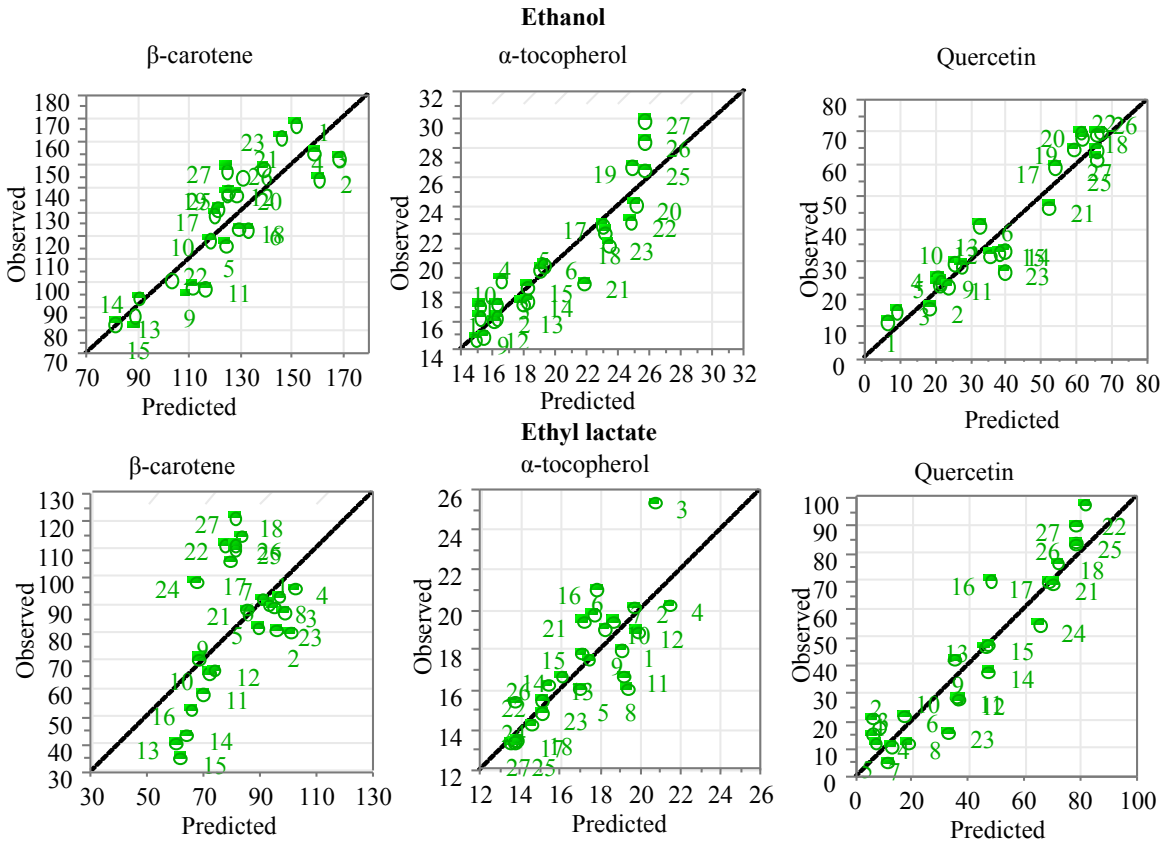

Figure S2. Plots show the linearity relationship between observed versus predicted values from the models for extraction of β-carotene, α-tocopherol, and quercetin using ethanol and ethyl lactate as a co-solvent.

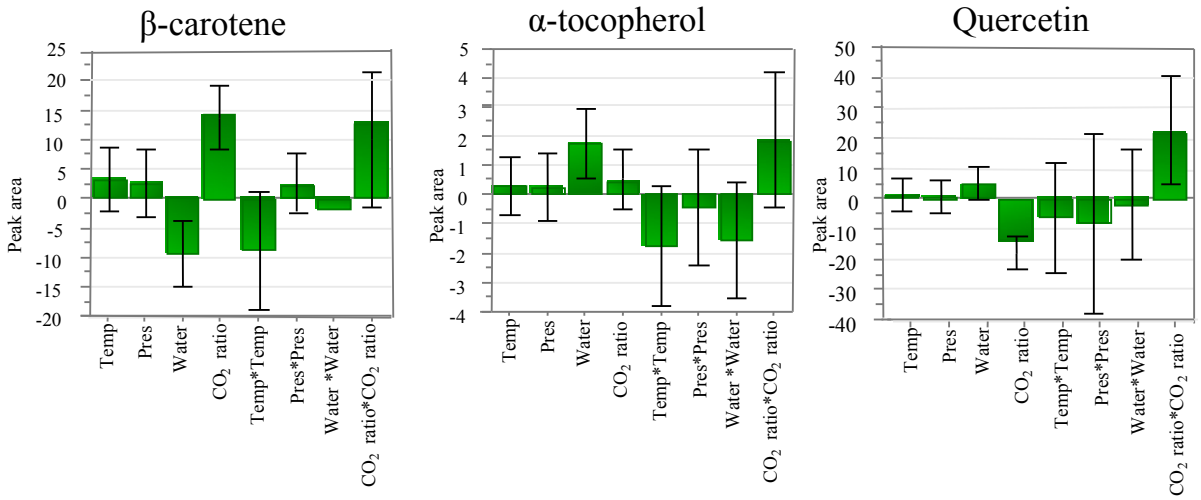

Figure S3. Coefficient plots from face centred central composite designs (FC-CCD) showing the influence of extraction parameter conditions on extraction recoveries of β-carotene and α-tocopherol using ethanol as a solvent, and quercetin using ethyl lactate.

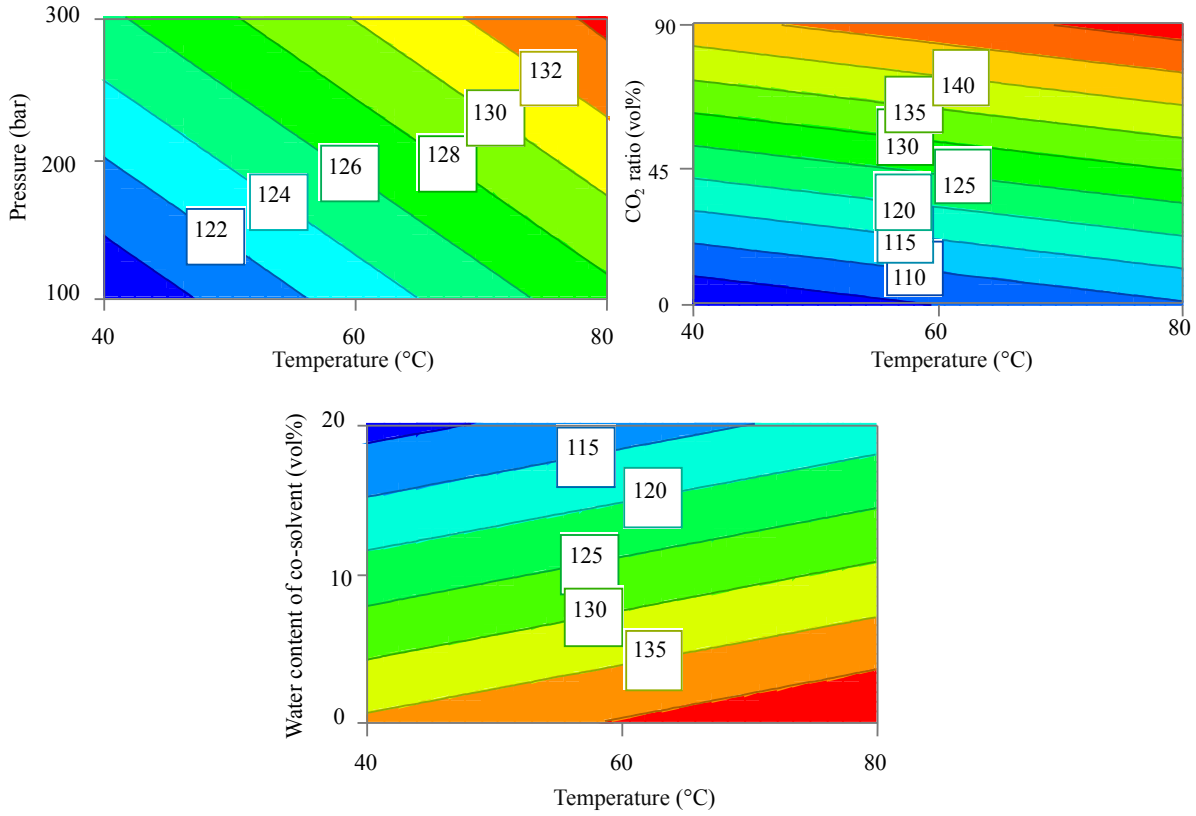

Figure S4. Contour plots show the direct and interactions influence of investigated parameters on the extracted amount of β-carotene (peak area, mAU). Ethanol is used as co-solvent.

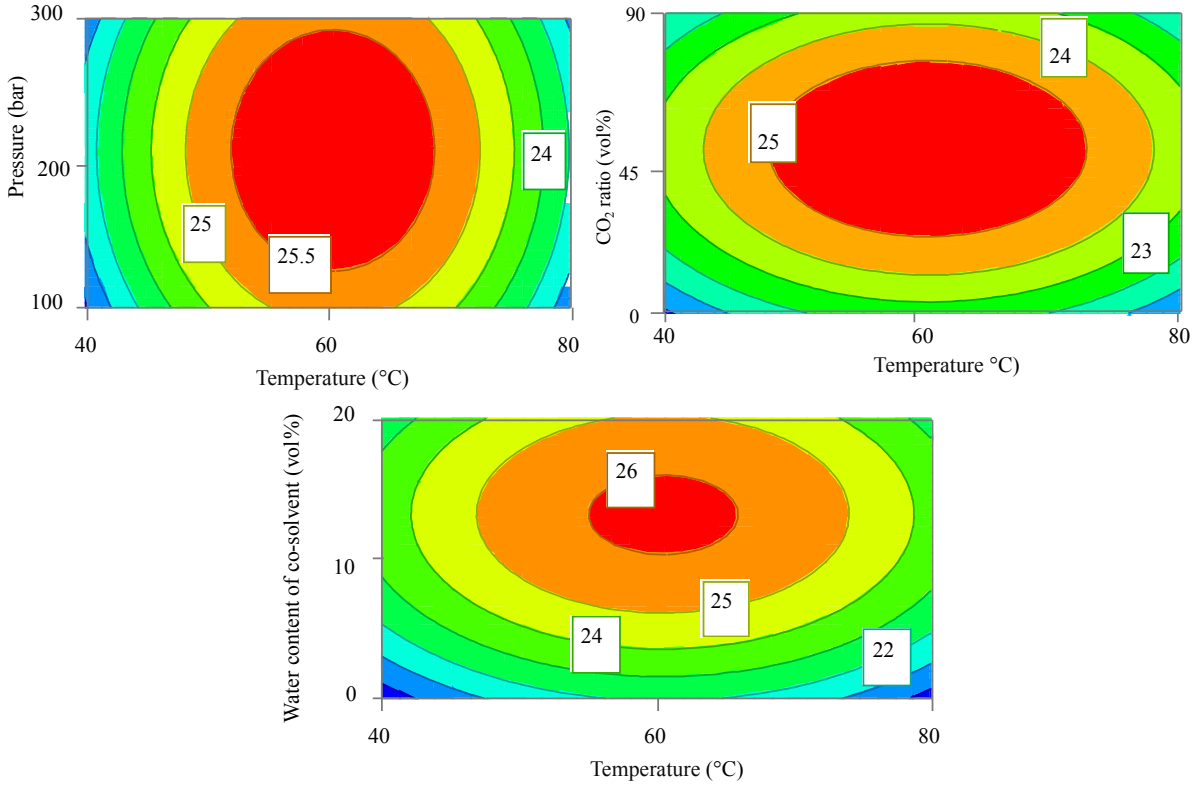

Figure S5. Contour plots show the direct and interactions influence of investigated parameters on the extracted amount of α-tocopherol (peak area, mAU). Ethanol is used as co-solvent.

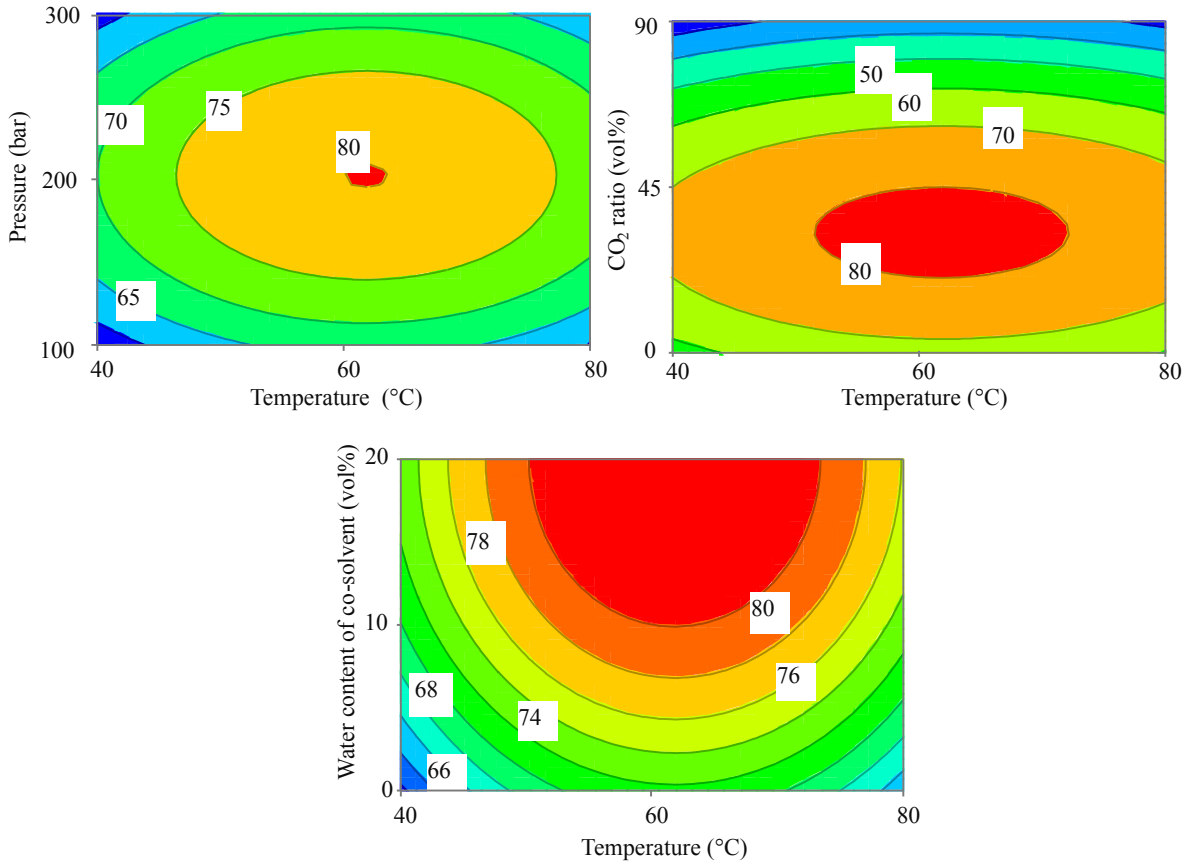

Figure S6. Contour plots show the direct and interactions influence of investigated parameters on the extracted amount of quercetin (peak area, mAU). Ethyl lactate is used as co-solvent.

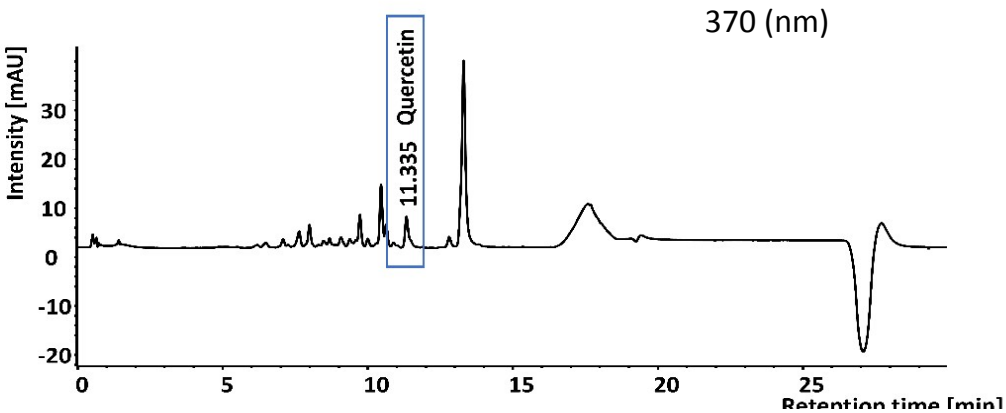

Figure S7. HPLC-DAD chromatogram at 370 nm from sea buckthorn pomace extract obtained by the developed method at the optimal conditions.

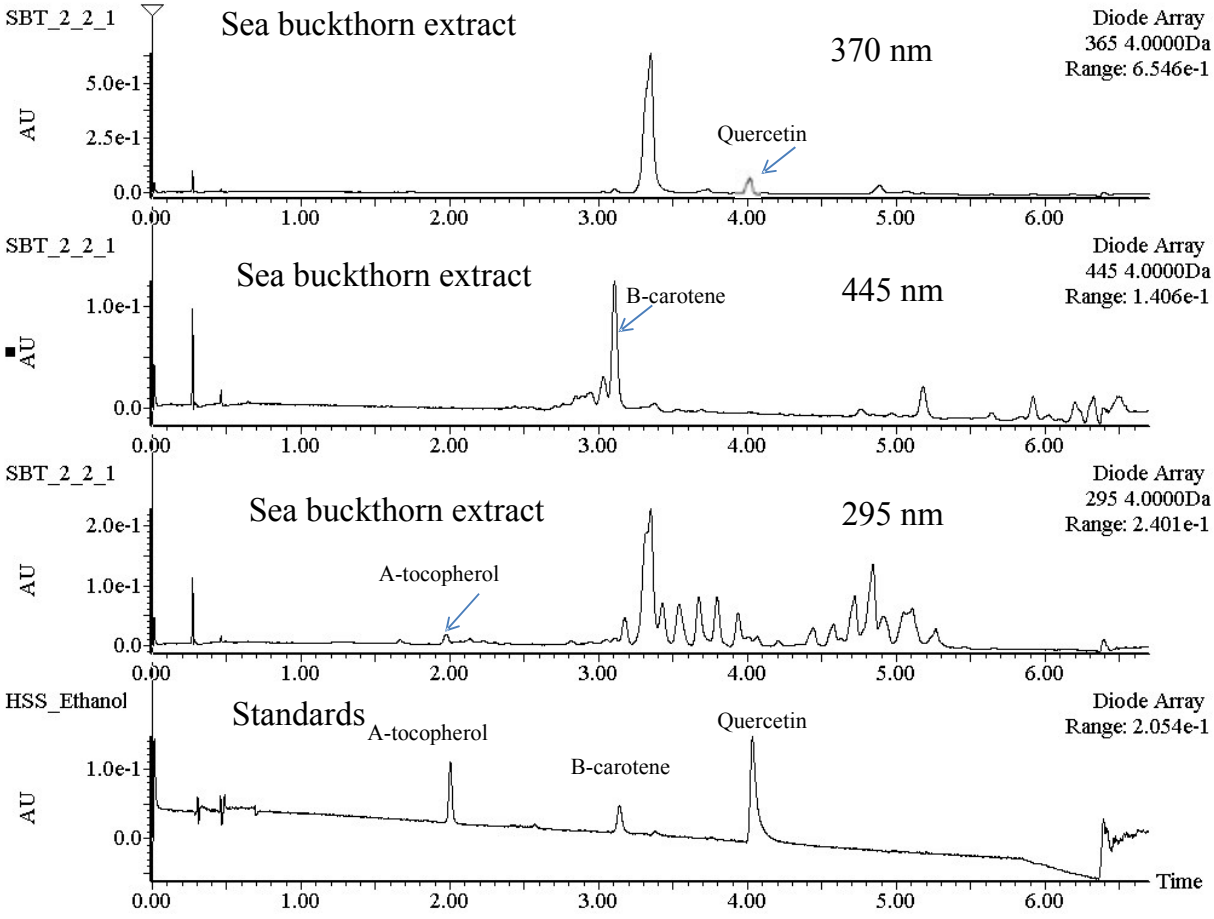

Figure S8. UHPSFC-DAD chromatogram for the standard of investigated compounds and sea buckthorn pomace extract obtained by the developed method at the optimal conditions. Extracted chromatograms are shown at 370 nm for quercetin, 445 nm for β-carotene and 290 nm for α-tocopherol.

Table S1. Calculation of the molar fractions of a ternary system consisting of water, ethanol, and CO_2 at different pressures and molar ratios of CO_2 to ethanol to water.

CO2 ratio 0 vol%

Water content (vol%)	*10*					
Total flow rate (ml/min)	*2,0*					
Compound	MW (g/mol)	Density (g/ml)	Volumetric flow rate (ml/min)	Mass flow rate (g/min)	Mole/min	**Mole ratio**
Water	18,015	1,000	0,2	0,2000	0,0111	**0,2648**
Ethanol	46,069	0,789	1,8	1,4202	0,0308	**0,7352**
				Total moles/min:	0,0419	

CO2 ratio 0 vol%

Water content (vol%)	*20*					
Total flow rate (ml/min)	*2,0*					
Compound	MW (g/mol)	Density (g/ml)	Volumetric flow rate (ml/min)	Mass flow rate (g/min)	Mole/min	**Mole ratio**
Water	18,015	1,000	0,4	0,4000	0,0222	**0,4476**
Ethanol	46,069	0,789	1,6	1,2624	0,0274	**0,5524**
				Total moles/min:	0,0496	

CO2 ratio 45 vol%

Total flow rate (ml/min)	*2,0*	*CO2 density calculated based on this pressure and 10°C (https://webbook.nist.gov/chemistry/fluid/)*				
Pressure (bar)	*100*					
Water content (vol%)	*0*					
CO2 ratio (volumetric)	*0,45*					
Water ratio (volumetric)	*0*					
Ethanol ratio (volumetric)	*0,6*					
Compound	MW (g/mol)	Density (g/ml)	Volumetric flow rate (ml/min)	Mass flow rate (g/min)	Mole/min	**Mole ratio**
Ethanol	46,069	0,789	1,1	0,8679	0,0188	**0,5002**
Carbon dioxide	44,009	0,921	0,9	0,8285	0,0188	**0,4998**
				Total moles/min:	0,0377	

CO2 ratio 45 vol%

Total flow rate (ml/min)	*2,0*	*CO2 density calculated based on this pressure and 10°C (https://webbook.nist.gov/chemistry/fluid/)*				
Pressure (bar)	*200*					
Water content (vol%)	*0*					
CO2 ratio (volumetric)	*0,45*					
Water ratio (volumetric)	*0*					
Ethanol ratio (volumetric)	*0,6*					
Compound	MW (g/mol)	Density (g/ml)	Volumetric flow rate (ml/min)	Mass flow rate (g/min)	Mole/min	**Mole ratio**
Ethanol	46,069	0,789	1,1	0,8679	0,0188	**0,4845**
Carbon dioxide	44,009	0,980	0,9	0,8823	0,0200	**0,5155**
				Total moles/min:	0,0389	

CO2 ratio 45 vol%

Total flow rate (ml/min)	*2,0*	*CO2 density calculated based on this pressure and 10°C (https://webbook.nist.gov/chemistry/fluid/)*				
Pressure (bar)	*300*					
Water content (vol%)	*0*					
CO2 ratio (volumetric)	*0,45*					
Water ratio (volumetric)	*0*					
Ethanol ratio (volumetric)	*0,6*					
Compound	MW (g/mol)	Density (g/ml)	Volumetric flow rate (ml/min)	Mass flow rate (g/min)	Mole/min	**Mole ratio**
Ethanol	46,069	0,789	1,1	0,8679	0,0188	**0,4746**
Carbon dioxide	44,009	1,020	0,9	0,9180	0,0209	**0,5254**
				Total moles/min:	0,0397	

CO2 ratio 45 vol%

Total flow rate (ml/min)	*2,0*	*CO2 density calculated based on this pressure and 10°C (https://webbook.nist.gov/chemistry/fluid/)*				
Pressure (bar)	*100*					
Water content (vol%)	*10*					
CO2 ratio (volumetric)	*0,450*					
Water ratio (volumetric)	*0,055*					
Ethanol ratio (volumetric)	*0,495*					
Compound	MW (g/mol)	Density (g/ml)	Volumetric flow rate (ml/min)	Mass flow rate (g/min)	Mole/min	**Mole ratio**
Water	18,015	1,000	0,110	0,110	0,0061	**0,1458**
Ethanol	46,069	0,789	0,990	0,781	0,0170	**0,4048**
Carbon dioxide	44,009	0,921	0,900	0,828	0,0188	**0,4494**
				Total moles/min:	0,0419	

Table S1. Cont.

CO2 ratio 45 vol%

Total flow rate (ml/min)	*2,0*
Pressure (bar)	*200*
Water content (vol%)	*10*
CO2 ratio (volumetric)	*0,450*
Water ratio (volumetric)	*0,055*
Ethanol ratio (volumetric)	*0,495*

CO2 density calculated based on this pressure and 10°C (https://webbook.nist.gov/chemistry/fluid/)

Compound	MW (g/mol)	Density (g/ml)	Volumetric flow rate (ml/min)	Mass flow rate (g/min)	Mole/min	**Mole ratio**
Water	18,015	1,000	0,110	0,110	0,0061	**0,1416**
Ethanol	46,069	0,789	0,990	0,781	0,0170	**0,3933**
Carbon dioxide	44,009	0,980	0,900	0,882	0,0200	**0,4650**
				Total moles/min:	0,0431	

CO2 ratio 45 vol%

Total flow rate (ml/min)	*2,0*
Pressure (bar)	*300*
Water content (vol%)	*10*
CO2 ratio (volumetric)	*0,450*
Water ratio (volumetric)	*0,055*
Ethanol ratio (volumetric)	*0,495*

CO2 density calculated based on this pressure and 10°C (https://webbook.nist.gov/chemistry/fluid/)

Compound	MW (g/mol)	Density (g/ml)	Volumetric flow rate (ml/min)	Mass flow rate (g/min)	Mole/min	**Mole ratio**
Water	18,015	1,000	0,110	0,110	0,0061	**0,1390**
Ethanol	46,069	0,789	0,990	0,781	0,0170	**0,3860**
Carbon dioxide	44,009	1,020	0,900	0,918	0,0209	**0,4749**
				Total moles/min:	0,0439	

CO2 ratio 45 vol%

Total flow rate (ml/min)	*2,0*
Pressure (bar)	*100*
Water content (vol%)	*20*
CO2 ratio (volumetric)	*0,450*
Water ratio (volumetric)	*0,110*
Ethanol ratio (volumetric)	*0,440*

CO2 density calculated based on this pressure and 10°C (https://webbook.nist.gov/chemistry/fluid/)

Compound	MW (g/mol)	Density (g/ml)	Volumetric flow rate (ml/min)	Mass flow rate (g/min)	Mole/min	**Mole ratio**
Water	18,015	1,000	0,220	0,220	0,0122	**0,2649**
Ethanol	46,069	0,789	0,880	0,694	0,0151	**0,3269**
Carbon dioxide	44,009	0,921	0,900	0,828	0,0188	**0,4083**
				Total moles/min:	0,0461	

CO2 ratio 45 vol%

Total flow rate (ml/min)	*2,0*
Pressure (bar)	*200*
Water content (vol%)	*20*
CO2 ratio (volumetric)	*0,450*
Water ratio (volumetric)	*0,110*
Ethanol ratio (volumetric)	*0,440*

CO2 density calculated based on this pressure and 10°C (https://webbook.nist.gov/chemistry/fluid/)

Compound	MW (g/mol)	Density (g/ml)	Volumetric flow rate (ml/min)	Mass flow rate (g/min)	Mole/min	**Mole ratio**
Water	18,015	1,000	0,220	0,220	0,0122	**0,2580**
Ethanol	46,069	0,789	0,880	0,694	0,0151	**0,3184**
Carbon dioxide	44,009	0,980	0,900	0,882	0,0200	**0,4236**
				Total moles/min:	0,0473	

CO2 ratio 45 vol%

Total flow rate (ml/min)	*2,0*
Pressure (bar)	*300*
Water content (vol%)	*20*
CO2 ratio (volumetric)	*0,450*
Water ratio (volumetric)	*0,110*
Ethanol ratio (volumetric)	*0,440*

CO2 density calculated based on this pressure and 10°C (https://webbook.nist.gov/chemistry/fluid/)

Compound	MW (g/mol)	Density (g/ml)	Volumetric flow rate (ml/min)	Mass flow rate (g/min)	Mole/min	**Mole ratio**
Water	18,015	1,000	0,220	0,220	0,0122	**0,2537**
Ethanol	46,069	0,789	0,880	0,694	0,0151	**0,3131**
Carbon dioxide	44,009	1,020	0,900	0,918	0,0209	**0,4333**
				Total moles/min:	0,0481	

Table S1. Cont.

CO2 ratio 90 vol%

Total flow rate (ml/min)	*2,0*	*CO2 density calculated based on this pressure and 10°C (https://webbook.nist.gov/chemistry/fluid/)*				
Pressure (bar)	*100*					
Water content (vol%)	*0*					
CO2 ratio (volumetric)	*0,900*					
Water ratio (volumetric)	*0,000*					
Ethanol ratio (volumetric)	*0,100*					
Compound	MW (g/mol)	Density (g/ml)	Volumetric flow rate (ml/min)	Mass flow rate (g/min)	Mole/min	**Mole ratio**
Ethanol	46,069	0,789	0,200	0,158	0,0034	**0,0834**
Carbon dioxide	44,009	0,921	1,800	1,657	0,0376	**0,9166**
				Total moles/min:	0,0411	

CO2 ratio 90 vol%

Total flow rate (ml/min)	*2,0*	*CO2 density calculated based on this pressure and 10°C (https://webbook.nist.gov/chemistry/fluid/)*				
Pressure (bar)	*200*					
Water content (vol%)	*0*					
CO2 ratio (volumetric)	*0,900*					
Water ratio (volumetric)	*0,000*					
Ethanol ratio (volumetric)	*0,100*					
Compound	MW (g/mol)	Density (g/ml)	Volumetric flow rate (ml/min)	Mass flow rate (g/min)	Mole/min	**Mole ratio**
Ethanol	46,069	0,789	0,200	0,158	0,0034	**0,0787**
Carbon dioxide	44,009	0,980	1,800	1,765	0,0401	**0,9213**
				Total moles/min:	0,0435	

CO2 ratio 90 vol%

Total flow rate (ml/min)	*2,0*	*CO2 density calculated based on this pressure and 10°C (https://webbook.nist.gov/chemistry/fluid/)*				
Pressure (bar)	*300*					
Water content (vol%)	*0*					
CO2 ratio (volumetric)	*0,900*					
Water ratio (volumetric)	*0,000*					
Ethanol ratio (volumetric)	*0,100*					
Compound	MW (g/mol)	Density (g/ml)	Volumetric flow rate (ml/min)	Mass flow rate (g/min)	Mole/min	**Mole ratio**
Ethanol	46,069	0,789	0,200	0,158	0,0034	**0,0759**
Carbon dioxide	44,009	1,020	1,800	1,836	0,0417	**0,9241**
				Total moles/min:	0,0451	

CO2 ratio 90 vol%

Total flow rate (ml/min)	*2,0*	*CO2 density calculated based on this pressure and 10°C (https://webbook.nist.gov/chemistry/fluid/)*				
Pressure (bar)	*100*					
Water content (vol%)	*10*					
CO2 ratio (volumetric)	*0,900*					
Water ratio (volumetric)	*0,010*					
Ethanol ratio (volumetric)	*0,090*					
Compound	MW (g/mol)	Density (g/ml)	Volumetric flow rate (ml/min)	Mass flow rate (g/min)	Mole/min	**Mole ratio**
Water	18,015	1,000	0,020	0,020	0,0011	**0,0265**
Ethanol	46,069	0,789	0,180	0,142	0,0031	**0,0737**
Carbon dioxide	44,009	0,921	1,800	1,657	0,0376	**0,8998**
				Total moles/min:	0,0418	

CO2 ratio 90 vol%

Total flow rate (ml/min)	*2,0*	*CO2 density calculated based on this pressure and 10°C (https://webbook.nist.gov/chemistry/fluid/)*				
Pressure (bar)	*200*					
Water content (vol%)	*10*					
CO2 ratio (volumetric)	*0,900*					
Water ratio (volumetric)	*0,010*					
Ethanol ratio (volumetric)	*0,090*					
Compound	MW (g/mol)	Density (g/ml)	Volumetric flow rate (ml/min)	Mass flow rate (g/min)	Mole/min	**Mole ratio**
Water	18,015	1,000	0,020	0,020	0,0011	**0,0251**
Ethanol	46,069	0,789	0,180	0,142	0,0031	**0,0696**
Carbon dioxide	44,009	0,980	1,800	1,765	0,0401	**0,9053**
				Total moles/min:	0,0443	

Table S1. Cont.

CO2 ratio 90 vol%

Total flow rate (ml/min)	*2,0*
Pressure (bar)	*300*
Water content (vol%)	*10*
CO2 ratio (volumetric)	*0,900*
Water ratio (volumetric)	*0,010*
Ethanol ratio (volumetric)	*0,090*

CO2 density calculated based on this pressure and 10°C (https://webbook.nist.gov/chemistry/fluid/)

Compound	MW (g/mol)	Density (g/ml)	Volumetric flow rate (ml/min)	Mass flow rate (g/min)	Mole/min	**Mole ratio**
Water	18,015	1,000	0,020	0,020	0,0011	**0,0242**
Ethanol	46,069	0,789	0,180	0,142	0,0031	**0,0671**
Carbon dioxide	44,009	1,020	1,800	1,836	0,0417	**0,9087**
				Total moles/min:	0,0459	

CO2 ratio 90 vol%

Total flow rate (ml/min)	*2,0*
Pressure (bar)	*100*
Water content (vol%)	*20*
CO2 ratio (volumetric)	*0,900*
Water ratio (volumetric)	*0,020*
Ethanol ratio (volumetric)	*0,080*

CO2 density calculated based on this pressure and 10°C (https://webbook.nist.gov/chemistry/fluid/)

Compound	MW (g/mol)	Density (g/ml)	Volumetric flow rate (ml/min)	Mass flow rate (g/min)	Mole/min	**Mole ratio**
Water	18,015	1,000	0,040	0,040	0,0022	**0,0521**
Ethanol	46,069	0,789	0,160	0,126	0,0027	**0,0643**
Carbon dioxide	44,009	0,921	1,800	1,657	0,0376	**0,8836**
				Total moles/min:	0,0426	

CO2 ratio 90 vol%

Total flow rate (ml/min)	*2,0*
Pressure (bar)	*200*
Water content (vol%)	*20*
CO2 ratio (volumetric)	*0,900*
Water ratio (volumetric)	*0,020*
Ethanol ratio (volumetric)	*0,080*

CO2 density calculated based on this pressure and 10°C (https://webbook.nist.gov/chemistry/fluid/)

Compound	MW (g/mol)	Density (g/ml)	Volumetric flow rate (ml/min)	Mass flow rate (g/min)	Mole/min	**Mole ratio**
Water	18,015	1,000	0,040	0,040	0,0022	**0,0493**
Ethanol	46,069	0,789	0,160	0,126	0,0027	**0,0608**
Carbon dioxide	44,009	0,980	1,800	1,765	0,0401	**0,8899**
				Total moles/min:	0,0451	

CO2 ratio 90 vol%

Total flow rate (ml/min)	*2,0*
Pressure (bar)	*300*
Water content (vol%)	*20*
CO2 ratio (volumetric)	*0,900*
Water ratio (volumetric)	*0,020*
Ethanol ratio (volumetric)	*0,080*

CO2 density calculated based on this pressure and 10°C (https://webbook.nist.gov/chemistry/fluid/)

Compound	MW (g/mol)	Density (g/ml)	Volumetric flow rate (ml/min)	Mass flow rate (g/min)	Mole/min	**Mole ratio**
Water	18,015	1,000	0,040	0,040	0,0022	**0,0476**
Ethanol	46,069	0,789	0,160	0,126	0,0027	**0,0587**
Carbon dioxide	44,009	1,020	1,800	1,836	0,0417	**0,8937**
				Total moles/min:	0,0467	

Table S1. Cont.

Optimum conditions, for b-carotene

CO2 ratio (vol%)	*90*
Total flow rate (ml/min)	*2,0*
Pressure (bar)	*300*
Water content (vol%)	*0*
CO2 ratio (volumetric)	*0,900*
Water ratio (volumetric)	*0,000*
Ethanol ratio (volumetric)	*0,100*

CO2 density calculated based on this pressure and 10°C (https://webbook.nist.gov/chemistry/fluid/)

Compound	MW (g/mol)	Density (g/ml)	Volumetric flow rate (ml/min)	Mass flow rate (g/min)	Mole/min	**Mole ratio**
Ethanol	46,069	0,789	0,200	0,158	0,0034	**0,0759**
Carbon dioxide	44,009	1,020	1,800	1,836	0,0417	**0,9241**
				Total moles/min:	0,0451	

Optimum conditions, for a-tocopherol

CO2 ratio (vol%)	*50*
Total flow rate (ml/min)	*2,0*
Pressure (bar)	*210*
Water content (vol%)	*12*
CO2 ratio (volumetric)	*0,500*
Water ratio (volumetric)	*0,060*
Ethanol ratio (volumetric)	*0,440*

CO2 density calculated based on this pressure and 10°C (https://webbook.nist.gov/chemistry/fluid/)

Compound	MW (g/mol)	Density (g/ml)	Volumetric flow rate (ml/min)	Mass flow rate (g/min)	Mole/min	**Mole ratio**
Water	18,015	1,000	0,120	0,120	0,0067	**0,1510**
Ethanol	46,069	0,789	0,880	0,694	0,0151	**0,3417**
Carbon dioxide	44,009	0,985	1,000	0,985	0,0224	**0,5073**
				Total moles/min:	0,0441	

Table S2. Experimental conditions of the DOE and the corresponding peak areas of investigated compounds using ethanol as an organic solvent.

Exp No	Exp Name	Run Order	Temperature °C	Pressure bar	Water content vol%	CO_2 ratio (vol%)	Peak Area (mAU)		
							β-carotene	α-tocopherol	Quercetin
1	N1	19	40	100	0	90	219	22	11
2	N2	26	80	100	0	90	147	18	16
3	N3	25	40	300	0	90	175	22	15
4	N4	21	80	300	0	90	185	22	26
5	N5	17	40	100	20	90	78	24	23
6	N6	10	80	100	20	90	98	21	41
7	N7	16	40	300	20	90	27	26	9
8	N8	13	80	300	20	90	87	26	36
9	N9	24	40	100	0	0	97	28	48
10	N10	20	80	100	0	0	109	29	59
11	N11	22	40	300	0	0	101	28	45
12	N12	27	80	300	0	0	103	33	58
13	N13	11	40	100	20	0	20	37	64
14	N14	18	80	100	20	0	17	31	67
15	N15	12	40	300	20	0	17	32	66
16	N16	15	80	300	20	0	38	36	72
17	N17	3	40	200	10	45	137	33	59
18	N18	1	80	200	10	45	229	21	68
19	N19	2	60	100	10	45	166	35	65
20	N20	4	60	300	10	45	132	18	70
21	N21	23	60	200	0	45	187	23	47
22	N22	14	60	200	20	45	154	32	70
23	N23	6	60	200	10	90	198	25	27
24	N24	9	60	200	10	0	94	38	70
25	N25	7	60	200	10	45	204	25	62
26	N26	8	60	200	10	45	170	27	69
27	N27	5	60	200	10	45	173	29	65

Table S3. Experimental conditions of DOE and the corresponding peak areas of investigated compounds using ethyl lactate as an organic solvent.

Exp No	Exp Name	Run Order	Temperature °C	Pressure bar	Water content vol%	CO_2 ratio (vol%)	Peak Area (mAU)		
							β-Carotene	α-Tocopherol	Quercetin
1	N1	19	40	100	0	90	92	18	15
2	N2	26	80	100	0	90	79	20	20
3	N3	25	40	300	0	90	87	25	17
4	N4	21	80	300	0	90	95	20	11
5	N5	17	40	100	20	90	81	16	4
6	N6	10	80	100	20	90	90	20	21
7	N7	16	40	300	20	90	92	19	10
8	N8	13	80	300	20	90	88	16	11
9	N9	24	40	100	0	0	70	17	41
10	N10	20	80	100	0	0	65	19	28
11	N11	22	40	300	0	0	58	17	28
12	N12	27	80	300	0	0	66	19	27
13	N13	11	40	100	20	0	39	16	46
14	N14	18	80	100	20	0	42	17	37
15	N15	12	40	300	20	0	35	18	46
16	N16	15	80	300	20	0	52	21	70
17	N17	3	40	200	10	45	105	13	69
18	N18	1	80	200	10	45	114	14	76
19	N19	2	60	100	10	45	127	13	29
20	N20	4	60	300	10	45	112	20	110
21	N21	23	60	200	0	45	88	19	69
22	N22	14	60	200	20	45	111	15	97
23	N23	6	60	200	10	90	80	15	15
24	N24	9	60	200	10	0	97	13	54
25	N25	7	60	200	10	45	109	13	83
26	N26	8	60	200	10	45	111	15	83
27	N27	5	60	200	10	45	121	13	89