

# Supercritical-Fluid Process Control to Functional Food Ingredient Development: *Lippia citriodora*

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**Supporting information Table 1A : Quantitation of individual compounds presents in *L. citriodora* supercritical extracts ( $\mu\text{g}$  of analyte/ g of dried extract). Value =  $X \pm \text{SD}$ .**

	SFE 1	SFE 2	SFE 3	SFE 4	SFE 5	SFE 6	SFE 7	SFE 8	SFE 9
<i>Iridioids</i>	4388 $\pm$ 185	1345 $\pm$ 68	5558 $\pm$ 369	899 $\pm$ 108	1460 $\pm$ 49	1706 $\pm$ 79	NQ	3708 $\pm$ 255	1357 $\pm$ 35
<i>Shanzhiside</i>	98 $\pm$ 19	NQ	240 $\pm$ 2	NQ	NQ	NQ	ND	NQ	NQ
<i>Gardoside</i>	2490 $\pm$ 71	805 $\pm$ 28	3057 $\pm$ 145	719 $\pm$ 57	1351 $\pm$ 37	1397 $\pm$ 34	NQ	2086 $\pm$ 78	1171 $\pm$ 3
<i>Theveside</i>	357 $\pm$ 22	NQ	475 $\pm$ 33	NQ	NQ	NQ	ND	111 $\pm$ 33	NQ
<i>Myxopyroside</i>	720 $\pm$ 2	433 $\pm$ 14	324 $\pm$ 41	180 $\pm$ 51	74 $\pm$ 4	268 $\pm$ 39	NQ	721 $\pm$ 74	181 $\pm$ 35
<i>Lamiidoside</i>	NQ	97 $\pm$ 23	215 $\pm$ 14	ND	NQ	NQ	ND	NQ	NQ
<i>Hydroxycampsid</i>	21 $\pm$ 14	NQ	239 $\pm$ 52	NQ	NQ	NQ	ND	49 $\pm$ 22	NQ
<i>Lippianoside B</i>	39 $\pm$ 2	NQ	248 $\pm$ 35	ND	NQ	NQ	ND	160 $\pm$ 17	NQ
<i>Durantioside I</i>	219 $\pm$ 41	NQ	467 $\pm$ 46	NQ	24 $\pm$ 3	NQ	ND	253 $\pm$ 18	NQ
<i>Manuleoside H</i>	429 $\pm$ 14	40 $\pm$ 3	293 $\pm$ 1	NQ	11 $\pm$ 5	41 $\pm$ 6	ND	328 $\pm$ 13	4.9 $\pm$ 0.4
<i>Phenylpropanoids</i>	5992 $\pm$ 97	2312 $\pm$ 136	28745 $\pm$ 1617	388 $\pm$ 20	4167 $\pm$ 265	1816 $\pm$ 121	184 $\pm$ 5	13318 $\pm$ 1216	6390 $\pm$ 249
<i>Verbascoside</i>	384 $\pm$ 21	NQ	655 $\pm$ 54	NQ	109 $\pm$ 9	12 $\pm$ 5	NQ	306 $\pm$ 28	61 $\pm$ 10
<i>Verbascoside</i>	3629 $\pm$ 30	1949 $\pm$ 99	21984 $\pm$ 1200	388 $\pm$ 20	3247 $\pm$ 214	1560 $\pm$ 73	184 $\pm$ 5	9764 $\pm$ 1042	5210 $\pm$ 195
<i>Lariciresinol glucopyranoside</i>	478 $\pm$ 11	147 $\pm$ 13	490 $\pm$ 39	NQ	133 $\pm$ 4	105 $\pm$ 16	NQ	449 $\pm$ 24	117 $\pm$ 13
<i>Isoverbascoside</i>	141 $\pm$ 8	NQ	1389 $\pm$ 129	NQ	81 $\pm$ 10	NQ	NQ	644.5 $\pm$ 0.1	207 $\pm$ 7
<i>Forsythoside A</i>	249 $\pm$ 7	NQ	1829 $\pm$ 107	NQ	144.1 $\pm$ 0.1	NQ	NQ	611 $\pm$ 18	366 $\pm$ 9
<i>Leucoseptoside A or isomer</i>	176 $\pm$ 2	NQ	990 $\pm$ 31	NQ	104 $\pm$ 8	NQ	NQ	445 $\pm$ 30	119 $\pm$ 1
<i>Leucoseptoside A or isomer</i>	NQ	NQ	NQ	ND	NQ	NQ	ND	NQ	NQ
<i>Martynoside or isomer</i>	604 $\pm$ 18	187 $\pm$ 14	974 $\pm$ 56	NQ	244 $\pm$ 14	107 $\pm$ 17	NQ	806 $\pm$ 46	255 $\pm$ 8
<i>Martynoside or isomer</i>	NQ	NQ	NQ	NQ	NQ	NQ	ND	NQ	NQ
<i>Osmanthisude B</i>	331.0 $\pm$ 0.6	29 $\pm$ 10	434 $\pm$ 1	NQ	105 $\pm$ 6	32 $\pm$ 10	NQ	293 $\pm$ 28	55 $\pm$ 6
<i>Flavonoids</i>	17833 $\pm$ 1099	17864 $\pm$ 617	18849 $\pm$ 1128	15061 $\pm$ 1010	14172 $\pm$ 518	17547 $\pm$ 500	11095 $\pm$ 983	12274 $\pm$ 841	23113 $\pm$ 355
<i>Methyl quercetin</i>	929 $\pm$ 63	958 $\pm$ 29	1111 $\pm$ 96	889 $\pm$ 52	885 $\pm$ 45	1117 $\pm$ 30	140 $\pm$ 9	718 $\pm$ 53	1044 $\pm$ 86
<i>Dimethyl Kaempferol</i>	5028 $\pm$ 285	4605 $\pm$ 59	5162 $\pm$ 147	3625 $\pm$ 124	3837 $\pm$ 261	4801 $\pm$ 114	1930 $\pm$ 156	3143 $\pm$ 18	6447 $\pm$ 48
<i>Dimethyl quercetin</i>	11876 $\pm$ 751	12301 $\pm$ 529	12576 $\pm$ 885	10547 $\pm$ 834	9450 $\pm$ 212	11629 $\pm$ 356	9025 $\pm$ 818	8413 $\pm$ 770	15622 $\pm$ 221
<i>Total</i>	28213 $\pm$ 1381	21521 $\pm$ 821	53152 $\pm$ 3111	16348 $\pm$ 1138	19799 $\pm$ 832	21069 $\pm$ 700	11279 $\pm$ 988	30183 $\pm$ 2312	30830 $\pm$ 639

\*NQ: Not quantified. Compound detected, but their concentration is between the detection and quantification limits. \*ND: Not detected. Compound concentration is below of detection limit.

**Supporting information Table 1B : Quantitation of individual compounds presents in *L. citriodora* supercritical extracts ( $\mu\text{g}$  of analyte/ g of extract). Value =  $X \pm \text{SD}$ .**

Condition Compound	SFE 10	SFE 11	SFE 12	SFE 13	SFE 14	SFE 15	SFE 16	SFE 17	SFE 18
<i>Iridioids</i>	<b>825 <math>\pm</math> 60</b>	<b>NQ</b>	<b>1416 <math>\pm</math> 101</b>	<b>7172 <math>\pm</math> 215</b>	<b>1715 <math>\pm</math> 101</b>	<b>1173 <math>\pm</math> 40</b>	<b>6663 <math>\pm</math> 436</b>	<b>3840 <math>\pm</math> 144</b>	<b>199 <math>\pm</math> 15</b>
<i>Shanzhiside</i>	NQ	ND	NQ	265 $\pm$ 25	NQ	NQ	147.8 $\pm$ 0.3	NQ	NQ
<i>Gardoside</i>	395 $\pm$ 15	NQ	976 $\pm$ 71	3136 $\pm$ 26	1019 $\pm$ 58	568 $\pm$ 34	2904 $\pm$ 188	1671 $\pm$ 2	19 $\pm$ 6
<i>Theveside</i>	NQ	ND	NQ	676 $\pm$ 11	NQ	NQ	505 $\pm$ 41	87 $\pm$ 17	NQ
<i>Myxopyroside</i>	408 $\pm$ 32	NQ	414 $\pm$ 24	680 $\pm$ 29	316 $\pm$ 10	574 $\pm$ 5	834 $\pm$ 59	1018 $\pm$ 22	180 $\pm$ 9
<i>Lamiidoside</i>	NQ	ND	NQ	326 $\pm$ 15	380 $\pm$ 33	NQ	299 $\pm$ 29	NQ	ND
<i>Hydroxycampsiside</i>	NQ	ND	NQ	354 $\pm$ 30	NQ	NQ	352 $\pm$ 50	21 $\pm$ 6	NQ
<i>Lippianoside B</i>	NQ	ND	NQ	503 $\pm$ 11	NQ	NQ	438 $\pm$ 25	187 $\pm$ 56	NQ
<i>Durantoside I</i>	NQ	ND	NQ	706 $\pm$ 20	NQ	NQ	619 $\pm$ 17	288 $\pm$ 18	NQ
<i>Manuleoside H</i>	22 $\pm$ 13	ND	26 $\pm$ 6	526 $\pm$ 48	NQ	31 $\pm$ 1	564 $\pm$ 27	568 $\pm$ 23	NQ
<i>Phenylpropanoids</i>	<b>1230 <math>\pm</math> 72</b>	<b>1531 <math>\pm</math> 77</b>	<b>3027 <math>\pm</math> 269</b>	<b>29429 <math>\pm</math> 1027</b>	<b>2620 <math>\pm</math> 86</b>	<b>9943 <math>\pm</math> 192</b>	<b>30448 <math>\pm</math> 1118</b>	<b>12739 <math>\pm</math> 491</b>	<b>2034 <math>\pm</math> 148</b>
<i>Verbascoside</i>	NQ	NQ	24.5 $\pm$ 0.7	747 $\pm$ 19	3.8 $\pm$ 0.1	NQ	641 $\pm$ 57	332 $\pm$ 19	NQ
<i>Verbascoside</i>	1079 $\pm$ 65	1467 $\pm$ 70	2489 $\pm$ 234	21847 $\pm$ 801	2219 $\pm$ 71	8537 $\pm$ 128	21908 $\pm$ 752	8631 $\pm$ 391	2034 $\pm$ 148
<i>Lariciresinol glucopyranoside</i>	56 $\pm$ 2	NQ	197 $\pm$ 6	734 $\pm$ 24	126 $\pm$ 0.6	99 $\pm$ 6	690 $\pm$ 24	562 $\pm$ 23	NQ
<i>Isoverbascoside</i>	NQ	NQ	13 $\pm$ 4	1837 $\pm$ 63	NQ	564 $\pm$ 13	1896 $\pm$ 17	737 $\pm$ 2	NQ
<i>Forsythoside A</i>	NQ	64 $\pm$ 7	9.6 $\pm$ 0.5	1267 $\pm$ 69	45 $\pm$ 3	94 $\pm$ 9	2415 $\pm$ 122	464 $\pm$ 46	NQ
<i>Leucoseptoside A or isomer</i>	NQ	NQ	5 $\pm$ 1	1078 $\pm$ 11	6 $\pm$ 1	215 $\pm$ 14	1058 $\pm$ 71	496 $\pm$ 0.4	NQ
<i>Leucoseptoside A or isomer</i>	NQ	NQ	NQ	NQ	NQ	NQ	NQ	NQ	ND
<i>Martynoside or isomer</i>	95 $\pm$ 5	NQ	249 $\pm$ 21	1301 $\pm$ 24	204 $\pm$ 9	355 $\pm$ 19	1237 $\pm$ 50	1070 $\pm$ 3	NQ
<i>Martynoside or isomer</i>	NQ	ND	NQ	NQ	NQ	NQ	NQ	NQ	NQ
<i>Osmanthisude B</i>	NQ	NQ	40 $\pm$ 2	618 $\pm$ 16	16 $\pm$ 1	79 $\pm$ 3	603 $\pm$ 27	447 $\pm$ 7	NQ
<i>Flavonoids</i>	<b>12070 <math>\pm</math> 716</b>	<b>8921 <math>\pm</math> 275</b>	<b>19452 <math>\pm</math> 1014</b>	<b>19675 <math>\pm</math> 854</b>	<b>19750 <math>\pm</math> 1385</b>	<b>13510 <math>\pm</math> 596</b>	<b>18171 <math>\pm</math> 1206</b>	<b>13336 <math>\pm</math> 917</b>	<b>12628 <math>\pm</math> 1000</b>
<i>Methyl quercetin</i>	626 $\pm$ 21	119 $\pm$ 10	1164 $\pm$ 59	1386 $\pm$ 19	1005 $\pm$ 45	502 $\pm$ 13	1588 $\pm$ 52	986 $\pm$ 97	617 $\pm$ 48
<i>Dimethyl Kaempferol</i>	2911 $\pm$ 182	1417 $\pm$ 89	5152 $\pm$ 247	5262 $\pm$ 316	5488 $\pm$ 124	3083 $\pm$ 295	5101 $\pm$ 373	3006 $\pm$ 63	2612 $\pm$ 110
<i>Dimethyl quercetin</i>	8533 $\pm$ 513	7385 $\pm$ 176	13136 $\pm$ 708	13027 $\pm$ 519	13257 $\pm$ 1216	9925 $\pm$ 288	11482 $\pm$ 781	9344 $\pm$ 757	9399 $\pm$ 842
<i>Total</i>	<b>14125 <math>\pm</math> 848</b>	<b>9182 <math>\pm</math> 352</b>	<b>23895 <math>\pm</math> 1384</b>	<b>56279 <math>\pm</math> 2096</b>	<b>24085 <math>\pm</math> 1572</b>	<b>24626 <math>\pm</math> 828</b>	<b>55282 <math>\pm</math> 2760</b>	<b>29915 <math>\pm</math> 1552</b>	<b>14861 <math>\pm</math> 1163</b>

\*NQ: Not quantified. Compound detected, but their concentration is between the detection and quantification limits. \*ND: Not detected. Compound concentration is below of detection limit.



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