

*Article*

# Essential oil-based nano-biopesticides: formulation and bioactivity against *Tribolium confusum*

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**Supplementary Table S1.** GC-MS analysis of the selected EOs.

<sup>a</sup> Calculated Linear Retention Index

Component	RLI <sup>a</sup>	<i>Pimpinella anisum</i> (%)	<i>Artemisia vulgaris</i> (%)	<i>Foeniculum vulgare</i> (%)	<i>Allium sativum</i> (%)	<i>Lavandula angustifolia</i> (%)	<i>Mentha piperita</i> (%)	<i>Rosmarinus officinalis</i> (%)	<i>Salvia officinalis</i> (%)
allyl methyl sulfide	699				0.57				
dimethyl disulfide	739				0.04				
hexan-2-one	797				0.03				
4-Methylthiazole	822				0.01				
allyl isopropyl sulfide	826				0.01				
furfural	832				0.01				
1,2-dithiolane	842				0.04				
unknown 1	851		0.23						
diallyl sulfide	857				11.09				
allyl propyl sulfide	872				0.04				
allyl methyl disulfide	914				2.3				
tricyclene	924		0.86						
α-thujene	928					0.05			0.21
methyl propyl disulfide	930				0.01				
α-pinene	934	0.51	0.61	2.63		0.91	1.98	11.28	21.2
2-ethoxythiazole	944				0.01				
methyl (E)-1-propenyl disulfide	947				0.13				
camphene	947		3.97	0.31		0.13		3.01	4.67
(E)-2-hexenal	952				0.03				
verbenene	953		0.18						
3H-1,2-dithiolene	957				0.13				
dimethyl trisulfide	967				0.02				
benzaldehyde	969				0.02				
sabinene	971	0.03	0.62	0.09		0.11	0.34	0.19	
β-pinene	974	0.16	0.4	0.98		0.23	0.99	4.01	0.23
3-(methylthio)-1-propanol	979				0.01				
1,2,4-trimethylbenzene	986		0.2						
β-myrcene	989	0.1	1.75	1.1		0.37	0.12	1.7	1.2



Component	LRI <sup>a</sup>	<i>Pimpinella anisum</i> (%)	<i>Artemisia vulgaris</i> (%)	<i>Foeniculum vulgare</i> (%)	<i>Allium sativum</i> (%)	<i>Lavandula angustifolia</i> (%)	<i>Mentha piperita</i> (%)	<i>Rosmarinus officinalis</i> (%)	<i>Salvia officinalis</i> (%)
allyl methyl trisulfide	1138				3.31				
menthone	1140						22.1		
pinocarvone	1149		0.19						
isomenthone	1149						8.75		
isoborneol	1153					0.41			
4-methyl-1,2,3-trithiolane	1156				0.99				
menthofurane	1156						1.1		
unknown 4	1158							0.22	
(-) neomenthol	1159						6.32		
borneol	1161		0.96			2.67		1.05	3.1
methyl (E)-1-propenyl trisulfide	1166				0.01				
(-) menthol	1170						37.67		
terpinene-4-ol	1171	0.18	0.69	0.17		1.74		0.42	
4,5-dimethyl-2-propylthiazole	1174				0.01				
hexyl butyrate	1181					0.19			
$\alpha$ -terpineol	1184	0.15				0.26	0.91	1.1	
estragol	1187	3.98		2.1					
3-vinyl-4H-1,2-dithiine	1189				0.03				
$\alpha$ -phellandrene epoxide	1195			0.21					
4H-1,2,3-trithiine	1200				0.02				
allicin (diallyl thiosulfinate)	1208				0.02				
2 vinyl-4H-1,3-dithiine	1215				0.03				
pulegone	1224						0.95		
4,5-dimethyl-2-butylthiazole	1226				0.08				
s-(+)-carvone	1226						1.66		
p-anisaldehyde	1235	0.41		0.72					
piperitone	1237						0.98		
(Z)-anethole	1243	0.21		0.3					
linalyl acetate	1252					36.31			

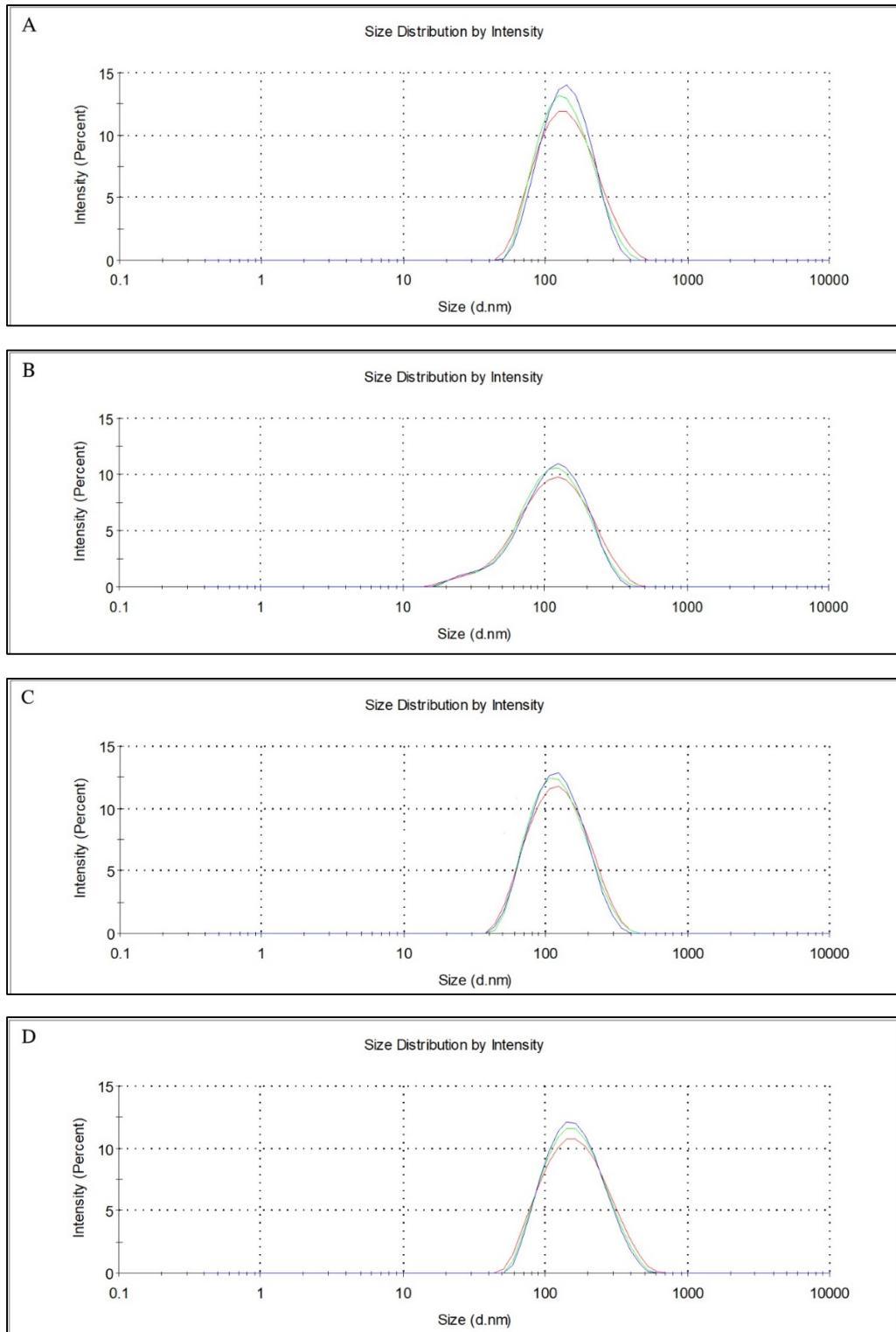
Component	LRI <sup>a</sup>	<i>Pimpinella anisum</i> (%)	<i>Artemisia vulgaris</i> (%)	<i>Foeniculum vulgare</i> (%)	<i>Allium sativum</i> (%)	<i>Lavandula angustifolia</i> (%)	<i>Mentha piperita</i> (%)	<i>Rosmarinus officinalis</i> (%)	<i>Salvia officinalis</i> (%)
chrysantenyl acetate	1256		4.97						
allyl isoproyl trisulfide	1266				0.05				
isomenthyl acetate	1270						0.21		
(E)-anethole	1274	87.11		42.13			3.93		
4-(hydroxymethyl)-1,2-dithiepane	1278				0.01				
(-)-bornyl acetate	1279						0.52	1.34	
isobornyl acetate	1279.5		0.48						
lavandulyl acetate	1282					1.17			
sabinyl acetate	1283		0.21						
4-methyl-1,2,5-trithiepane	1285				0.05				
diallyl trisulfide	1303				22.7				
allyl propyl trisulfide	1314				0.11				
(E)-3,5-diethyl-1,2,4-trithiolane	1342				0.04				
δ-elemene	1343					0.03			
eugenol	1346			1.08					
allyl methyl tetrasulfide	1357				0.01				
5-methyl-1,2,3,4-tetrathiane	1367				0.03				
geranyl acetate	1371					0.23			
(Z)-3,5-diethyl-1,2,4-trithiolane	1374				0.26				
eucarvone	1376		1.01						
α-ylangene	1377						0.04		
(-)-α-copaene	1381					0.12			
2-heptyl thiophene	1381				1.39				
α-copaene	1382	0.07	0.58					0.13	
daucene	1386					0.06			
β-bourbonene	1389						0.27		
β-elemene	1395						0.64		
unknown 5	1407					0.05			
longifolene	1412							0.12	0.67

Component	LRI <sup>a</sup>	<i>Pimpinella anisum</i> (%)	<i>Artemisia vulgaris</i> (%)	<i>Foeniculum vulgare</i> (%)	<i>Allium sativum</i> (%)	<i>Lavandula angustifolia</i> (%)	<i>Mentha piperita</i> (%)	<i>Rosmarinus officinalis</i> (%)	<i>Salvia officinalis</i> (%)
$\alpha$ -cedrene	1418					0.02			
unknown 6	1419	0.04			0.2				
$\beta$ -caryophyllene	1425	0.21	0.7			2.21	1.74	2.1	3.9
3,6-dimethyl-1,2,5-trithiepane	1428				0.01				
$\beta$ -cubebene	1433						0.5		
unknown 7	1439					0.3			
$\alpha$ -bergamotene	1439	0.25	0.19						
4-ethyl-2,3,5-trithia-6-octene	1444				0.36				
$\beta$ -farnesene	1454	0.01				0.61			
$\alpha$ -humulene	1459					0.32	0.12		3.99
4,6-dimethyl-1,2,5-trithiepane	1460				0.06				
$\alpha$ -elemene	1474					0.05			
$\gamma$ -muurolene	1479					0.06	0.03		
germacrene D	1484		0.8			0.32	0.39		
$\delta$ -guaiene	1485								0.04
germacrene B	1501		0.04				0.13		
$\alpha$ -farnesene	1503	0.02							
$\alpha$ -bisabolene	1508	0.07				0.13			
$\beta$ -cadinene	1511							0.01	
$\gamma$ -muurolene	1515	0.01							
$\gamma$ -cadinene	1515					0.14	0.06		
$\beta$ -sesquiphellandrene	1521					0.13			
$\delta$ -cadinene	1524	0.05					0.17	0.19	
diallyl tetrasulfide	1544				13.17				
(E)-nerolidol	1558	0.06							
allyl methyl pentasulfide	1573				0.01				
(+)-spathulenol	1579						0.03		
caryophyllene oxyde	1582					0.1	0.18	0.16	0.1
7-methyl-4,5,8-trithia-1,10-undecadiene	1583				0.34				

Component	LRI <sup>a</sup>	<i>Pimpinella anisum</i> (%)	<i>Artemisia vulgaris</i> (%)	<i>Foeniculum vulgare</i> (%)	<i>Allium sativum</i> (%)	<i>Lavandula angustifolia</i> (%)	<i>Mentha piperita</i> (%)	<i>Rosmarinus officinalis</i> (%)	<i>Salvia officinalis</i> (%)
4-ethyl-6-methyl-1,2,3,5-tetrathiolane	1588				0.05				
6-methyl-4,5,8-trithia-1,10-undecadiene	1592				1.19				
2-thiopheneacetamide, N-propyl-	1622				0.05				
4-methyl-1,2,3,5,6-pentathiepane	1649				0.22				
t-muurolol	1654	0.06							
6-ethyl-4,5,7,8-tetrathianonane	1658				0.15				
2-thiopheneacetamide, N-isobutyl-	1663				0.21				
foeniculin	1666	2.01		0.51					
α-bisabolol	1678					0.1			
hexathiepane	1680				0.24				
diallyl pentasulfide	1755				0.5				
allyl methyl hexasulfide	1781				0.51				
8-methyl-4,5,6,9-tetrathia-1,11-dodecadiene	1815				5.89				
diallyl hexasulfide	1897				0.26				
allyl methyl heptasulfide	1922				0.16				
2-methyl-1,3-benzothiazole	1957				0.36				
5-ethyl-7-pentyl-1,2,3,4,6-pentathiepane	2005				0.32				
cyclooctasulfur	2044				0.08				
9-methyl-4,5,6,7,10-pentathia-1,12-tridecadiene	2051				0.9				

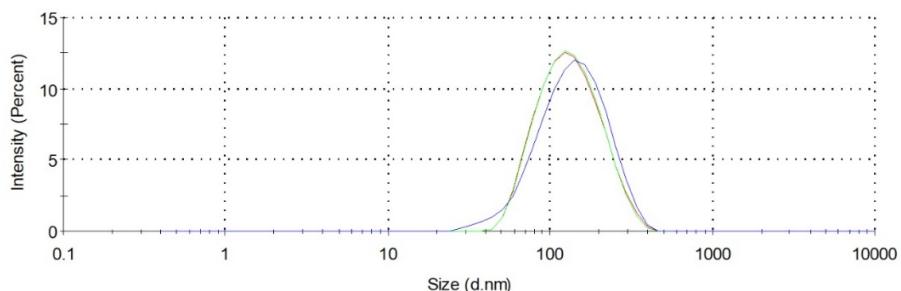
Component	LRI <sup>a</sup>	<i>Pimpinella anisum</i> (%)	<i>Artemisia vulgaris</i> (%)	<i>Foeniculum vulgare</i> (%)	<i>Allium sativum</i> (%)	<i>Lavandula angustifolia</i> (%)	<i>Mentha piperita</i> (%)	<i>Rosmarinus officinalis</i> (%)	<i>Salvia officinalis</i> (%)
8-methyl-4,5,6,7,10-pentathia-1,12-tridecadiene	2056				0.68				
<b>Monoterpene</b>		<b>2.96</b>	<b>11.33</b>	<b>44.28</b>		<b>3.11</b>	<b>11.16</b>	<b>28.47</b>	<b>32.66</b>
Oxygenated monoterpene		2.31	84.52	8.88		87.88	80.65	68.52	58.64
<b>Sesquiterpene</b>		<b>0.69</b>	<b>2.31</b>			<b>4.43</b>	<b>4.05</b>	<b>2.59</b>	<b>8.6</b>
Oxygenated sesquiterpene		0.12				0.2	0.21	0.16	0.1
<b>Phenylpropene</b>		<b>93.72</b>		<b>46.84</b>			<b>3.93</b>		
Ester						<b>0.19</b>			
<b>Sulphur compounds</b>					<b>99.8</b>				
<b>Aromatic hydrocarbon</b>			<b>0.45</b>						
<b>Total identified</b>		<b>99.80</b>	<b>98.61</b>	<b>100.00</b>	<b>99.80</b>	<b>95.81</b>	<b>100.00</b>	<b>99.74</b>	<b>100.00</b>

**Supplementary Figure S1.** Dimensional values of nanoemulsions containing anise(A), artemisia(B), fennel(C), garlic(D), lavender(E), mint(F), rosemary(G) or sage(H) EO.



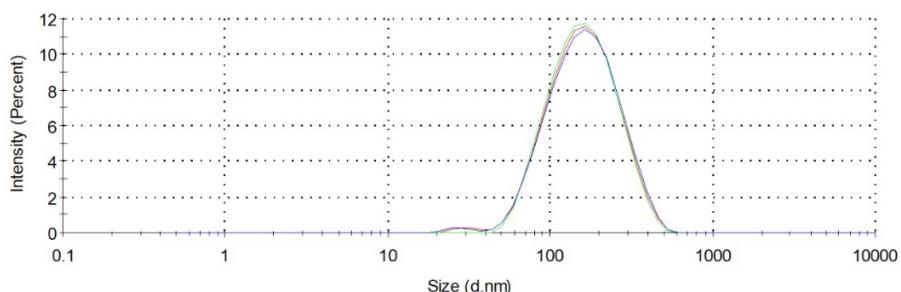
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Size Distribution by Intensity



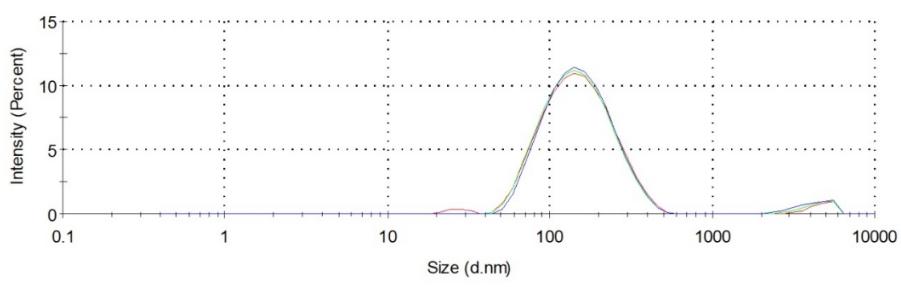
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Size Distribution by Intensity



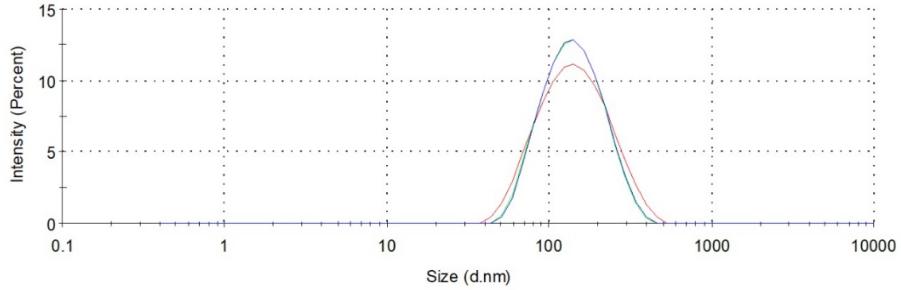
G

Size Distribution by Intensity

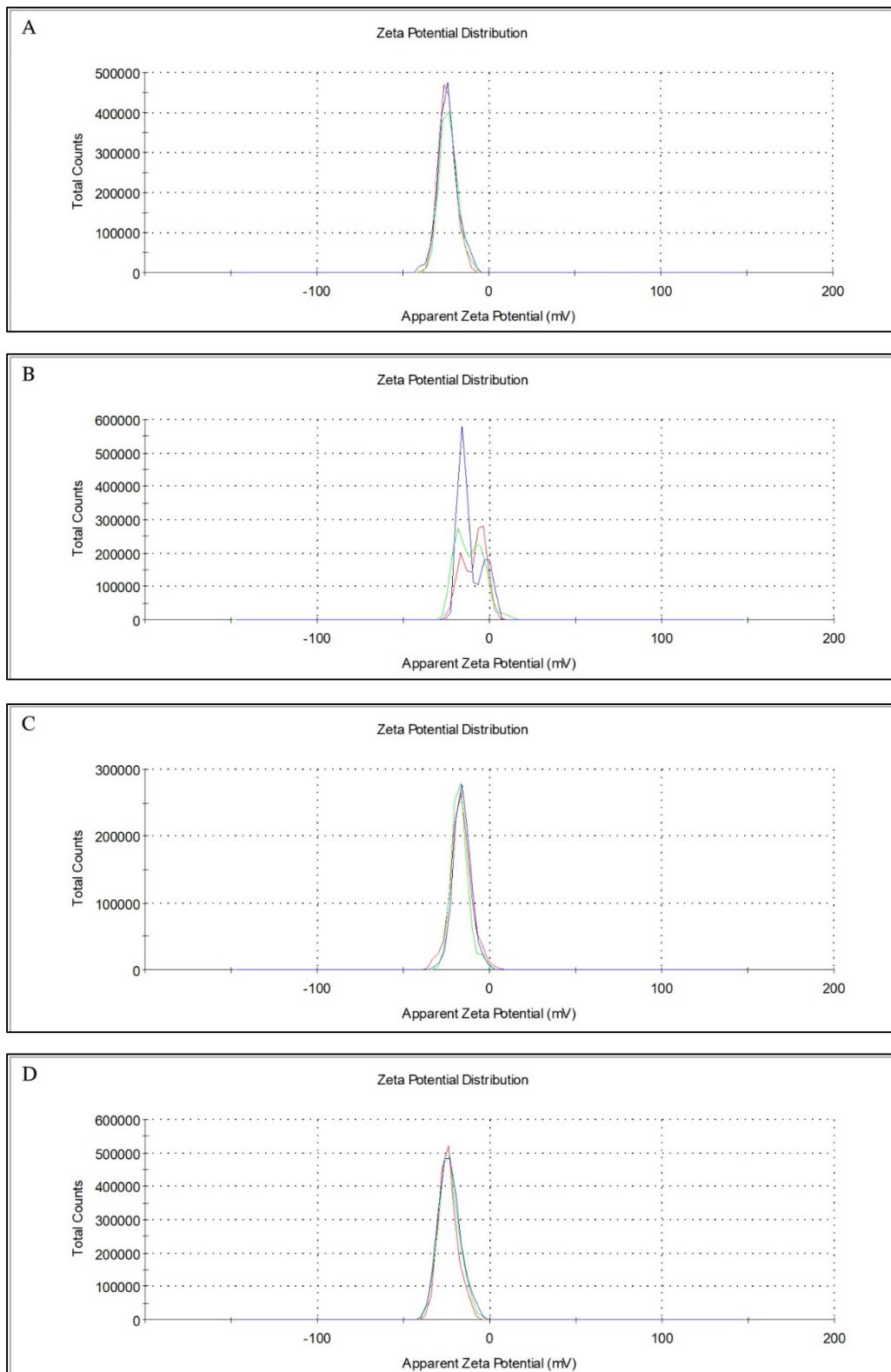


H

Size Distribution by Intensity

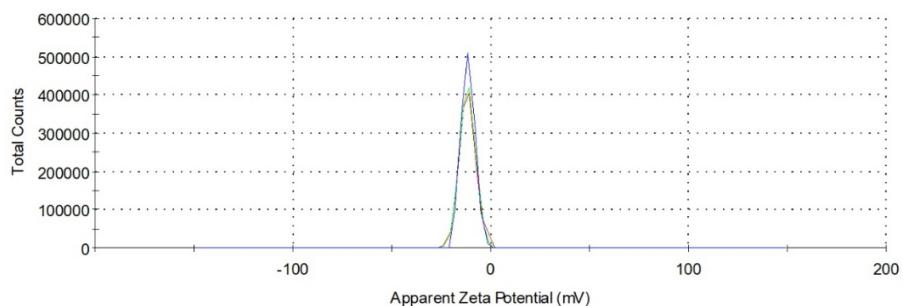


**Supplementary Figure S2.**  $\zeta$  potential values of nanoemulsions containing anise(A), artemisia(B), fennel(C), garlic(D), lavender(E), mint(F), rosemary(G) or sage(H) EO.



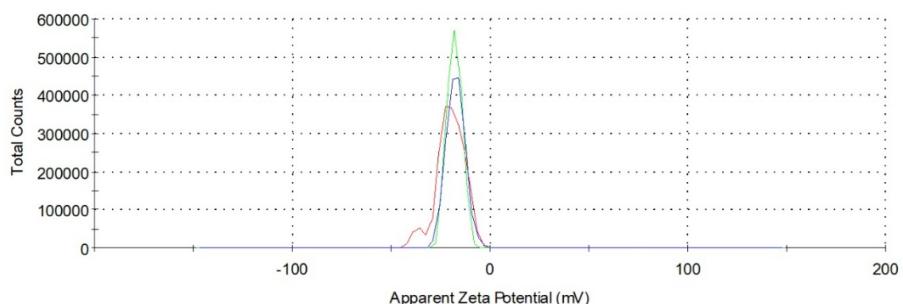
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Zeta Potential Distribution



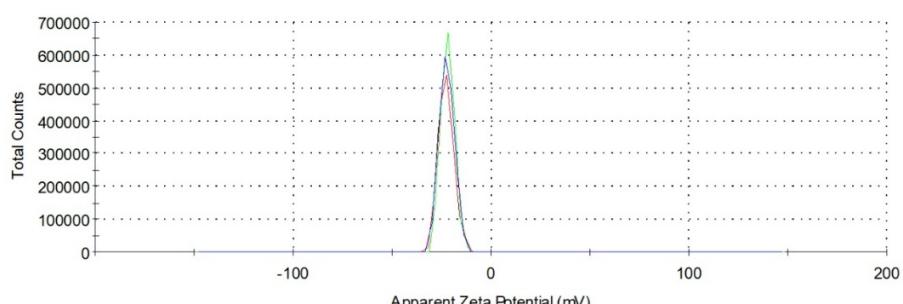
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Zeta Potential Distribution



G

Zeta Potential Distribution



H

Zeta Potential Distribution

