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1 *Journal of the Institute of Brewing*
2 ***Influences of distillation parameters on extraction of *Juniperus communis* L in vapour***
3 ***infused gin***

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8

9 **Abstract:**

10 The influence of distillation parameters on the extraction of specific flavour compounds
11 from the main gin botanical *Juniperus communis* L. using vapour infusion was investigated.
12 The experiment aimed to investigate an optimum mix of conditions to extract the highest
13 levels of certain compounds using a 3³-factorial design. Using solid-phase microextraction
14 and gas chromatography coupled mass spectrometry, quantification was carried out
15 measuring 9 common flavour compounds. The greatest influence was the botanical ratio,
16 whilst the least influential parameter was ethanol concentration for all compounds except
17 terpinen-4-ol. Density functional theory calculations of quantitative structure–activity
18 relationship properties for the flavour compounds were also undertaken to test for
19 molecular influence on solvent extraction. The quantitative structure–activity relationship
20 model showed the highest dipole moment for terpinen-4-ol and the lowest for γ -terpinene.

21

22

23 **Keywords: Distillation, Extraction, Flavour, Gin, *Juniperus communis* L.,**
24 **Quantitative structure–activity relationship**

25

Introduction

Gin is a distilled alcoholic beverage produced by the flavouring of neutral alcohol. Different definitions on the production of gin exist, with a common difference being the defined ethanol concentration of the neutral alcohol used, although all agree the predominant flavour should be of juniper (1–3). Here, the influence of certain distillation parameters on the extraction of volatile compounds in gin distillation was considered, and distillations using solely *Juniper communis* L. were conducted to gain a better understanding of the extraction behaviour of certain volatile compounds.

Gin distillation

Gin distillation or flavouring is achieved by two common methods of extraction: steep infusion or vapour infusion (4). The objective of gin distillation is a balance of extracting the right amount of desirable flavour without an excessive extraction of essential oils. Nevertheless, production of gin with solely essences or essential oils (EO), then commonly described as compound gin, is also possible (5). The process of gin production involves the extraction of flavour compounds from the botanicals into an ethanol-water mixture, vapourisation of the extract, reflux of the mixture – which varies based on the equipment design, and condensation of vapour into liquid again. The most common and traditional way to distil gin is steep infusion, where a mix of juniper berries and other botanicals are steeped in neutral alcohol or grain neutral spirit (GNS) usually diluted to between 45–65% vol., which can be additionally macerated for a specific period of time and temperature (6, 7). The mixture is then distilled, commonly in copper pot stills, and the condensed liquid is separated into fractions called foreshots, hearts (gin), and tails (8). Investigations on gin distillation under vacuum has also been conducted showing differences in concentrations comparing vacuum distillation with traditional distillation resulting in a 5-fold higher concentration of α -pinene, α -phellandrene, β -caryophyllene and β -myrcene using the traditional method as well as difference in flavour perception (9). In vapour infusion, botanicals are placed into a basket or chamber, which can either be placed inside the pot still or externally before the condenser. A most current comparison of the two methods showed that the vapour extraction resulted in a higher extraction monoterpenes than in steep infusion (10). The mix of ethanol-water vapour passes through the plant material and extracts the desired flavour compounds (4), characterised by:

- 1) Entrance of the solvent into the plant matrix
- 2) Solubilization of compound/ thermal oil exudation from the plant
- 3) Transport of compounds to the outside
- 4) Vapour-liquid equilibrium at the surface layer
- 5) Vapourisation (11–14)

Most available research on extraction behaviour only investigates the extraction with pure water via steam distillation (15–17) or hydro distillation (11, 18, 19).

Essential oil extraction and solvent polarity

Monoterpenes, sesquiterpenes and diterpenes are the most common constituents of essential oils from juniper. Studies that have investigated the flavour of juniper have either

67 considered these compounds in the form of essential oils (EO) (20, 21) or have specifically
68 focussed on their impact on the flavour of gin itself (22–24). Steam and hydro distillation
69 are both well-established methods used to isolate EO and are comparable to gin distillation.
70 It is worth mentioning that during flavouring of the base spirit, the most efficient essential
71 oil extraction is not necessarily the aim, but rather extracting the more fragrant and
72 appropriate flavour compounds to achieve a balanced product. This, however, prompts the
73 question of the extraction dynamics of such flavour constituents. Modelling of equivalent
74 extraction kinetics in cases of distillations of essential oils using solvents have been
75 conducted in literature for supercritical fluid extraction (25, 26), Soxhlet (27),
76 distillation-solvent extraction (SDE) (24) and hydroalcoholic extraction (28). The usage of
77 solvents in general, and specifically ethanol is of interest, as water and ethanol form a
78 non-ideal mixture. Whilst both are considered polar solvents, water (1.000) is more polar
79 than ethanol (0.654) (29). In studies of solvatochromic shifts, Lerf and Suppan (30)
80 concluded that micellar solvation clusters and solvent exchange equilibria control the
81 solvation behaviour and are defined by diffusion coefficients, the ability of the solute to
82 hydrogen bond, the dielectric constant and other enrichment parameters. Further work by
83 Zhong and Patel (31) modelled cluster sizes, showing that in ethanol-water mixtures
84 electrostatic polarization effects are one of the key factors, which are in turn influenced by
85 the molecular structure of the solute. By calculating some of these molecular characteristics
86 for flavour and aroma compounds such as polarisability, dielectric constants and
87 hydrogen-bond ability, it may be possible to predict their extraction in certain solvent
88 mixtures. This research investigates the optimal conditions to extract the highest levels of
89 certain compounds in gin distillation, by changing solute amount, concentration and charge
90 of the solvent.

91

92

Material and Methods

93 DFT Modelling of Key Aroma Compounds

94 Density Functional Theory (DFT) modelling of flavour and aroma compounds was
95 computed with Wavefunction Inc. Spartan 10 version 1.1.0, using a B3LYP hybrid
96 functional and a 6-311G* basis set (32–34).

97

98 Plant material

99 Ripe Macedonian *Juniperus communis* L. berries were purchased from Beacon
100 Commodities Ltd (Sussex, UK) in 2018 and were stored in a refrigerator (~8°C). The
101 essential oil content was measured at 1.217% w/w, obtained via hydro distillation with a
102 Clevenger type apparatus (19).

103

104 Grain neutral Spirit

105 The grain neutral spirit (wheat) used was purchased from Haymankimia (Witham, UK) at
106 96.2% ABV. The spirit was diluted with distilled water.

107
108
109

110 **Chemicals and analytics**

111 The constituents of juniper berries (21, 24) were chosen for quantification (Table 1). The
112 method of quantification used an AOC 5000 auto sampler coupled to a GCMS-QP2010
113 Ultra (Shimadzu, Kyoto, Japan). The sampling was carried out using a 65 µm PDMS/DVB
114 solid phase microextraction fibre (SPME) from Supelco (Bellefonte, USA) and for the
115 separation a HP5MS (30m x 0.25mm x 0.25µm) column from Agilent (Santa Clara, USA)
116 was used (10). Each sample was diluted to 8% ABV using distilled water. The internal
117 standard (3-octanol) at a final concentration of 9.48 mg/L was to the total volume of 5 mL
118 of diluted sample. The headspace vials (75.5x22.5) used were 20 mL with magnetic caps.

119

120 **Experimental design**

121 The experiment design was a 3³ - factorial response surface methodology resulting in 27
122 individual distillations. The functional dependence between the extracted amounts of the
123 chemical compounds listed in Table 1 was investigated as a response to the independent
124 variables ethanol (solvent) concentration of the charge, weight of juniper berries (solute)
125 and different bulk charges. The ethanol concentration ranged from 45% ABV, to 65%
126 ABV, the weight of the botanicals or ratio ranged from 2 g/L (-1), 4 g/L (0) to 6 g/L (+1)
127 and the bulk charge ranged from 200 mL to 400 mL (Table 2).

128

129 Equation 1 describes the interaction of the juniper berry extraction response to the three
130 main parameters.

131

$$132 \quad y = \beta_0 + \sum_{i=1}^3 \beta_i x_i + \sum_{i=1}^3 \beta_{ii} x_i^2 + \sum_{i=1}^2 \sum_{j=i+1}^3 \beta_{ij} x_i x_j \quad (1)$$

133

134 The variable y was defined as the concentration of each terpene, β_0 being a constant
135 coefficient, while β_i , β_{ii} , and β_{ij} denoted the regression coefficients of linear, square, and
136 interaction influences, respectively, and x_i and x_j were the independent variables affecting
137 the response values (35). The optimum distillation condition, defined as the combination of
138 parameters giving the highest extracted concentrations of the terpenes, was done in
139 triplicate using the same process described as follows.

140 **Distillation Process and Equipment**

141 The distillation setup was performed in laboratory glassware as described previously (10).
142 The experimental setup consisted of a glass extraction funnel with a vapour chamber and a
143 support for the botanical material, copper gauze, 100 mesh (0.11mm) Figure 1.

144

145

146 The distillate was collected into volumetric flasks to a volume of half the charge
147 (100/150/200 mL). Cooling water supply was maintained at a flowrate of 10 ml/s at 12.5
148 ± 2.5 °C. Additionally, the botanical funnel was drained (5.5 mL ± 0.5 mL) every 10 minutes
149 after the first liquid was obtained. All distillates were stored at 4 °C after distillation until
150 further usage.

151

152 **Sensory panel and Quantitative Descriptive Analysis**

153 A sensory panel of 20 participants was provided with 6 samples of gin distillates. Each
154 panellist was introduced in a pre-session to 9 flavour compounds presented in an opaque
155 glass filled with cotton wool, present in a concentration 3 times the amount of the highest
156 measured concentration of α -pinene to guarantee recognition. A standardised procedure of
157 sensory evaluation was introduced to each panellist and samples using the distillation
158 parameters of 2 g/L, 4 g/L and 6 g/L with an initial charge of 400 mL at 45% ABV and
159 65% ABV ethanol were assessed.

160 The sensory sessions were performed according to the BS ISO 6658:2017 (36) and BS
161 ISO 4121:2003 (37). Each sample was diluted to 20% ABV ethanol and presented in a
162 randomized order and blinding codes using sensory software (Compusense Inc., Ontario,
163 Canada). Panellists were asked to rate each descriptor (Table 5) from 0-9, with 0 being
164 “absent” and 9 “present” in a high concentration.

165

166

Results & Discussion

167 **DFT Modelling of Key Aroma and Flavour Compounds**

168 The solubilisation of any chemical compound is partly governed by the balance between the
169 polarity of the molecule (Figure 2) and that of the extracting solvent.

170

171 Increasing the still charge ethanol concentration, concurrently reduces the polarity of the
172 solvent mixture, albeit non-ideally, as the proportion of water decreases (29, 38, 39).
173 Quantitative structure–activity relationships (QSAR) including dipole moment,
174 polarisability and electrostatic surface potential (ESP) of key juniper extractives (Table 3)
175 were calculated by DFT methods. These molecular properties were correlated with solvent
176 extraction performance to predict the influence of solvent interactions, including hydrogen
177 bonding and van der Waal’s forces in the distillation of the gin samples.

178

179 The magnitude of charge separation, or dipole moment, is largest for compounds that
180 contain electronegative heteroatoms (i.e. oxygen), which pull electron density to create a
181 charge gradient with the electron rich and poor areas key for solvent interactions. The
182 calculations show that the oxygenated monoterpene terpinen-4-ol has the highest value of
183 those investigated (Figure 2). The electrostatic surface potential (ESP) can be used to
184 predict hydrogen-bonding potential by highlighting the more hydrogen bond acidic sites,
185 for example with terpinene-4-ol the more negative red area on the oxygen is the likely
186 hydrogen bond acceptor site, and the more positive blue area (the hydroxyl proton) is the

187 hydrogen bond donor site and by their magnitude consequently predict the strength of this
188 interaction. By contrast γ -terpinene has the lowest observed dipole value (0.07 debye),
189 with a combination of only carbon atoms with similar electronegativity and no connected
190 π - π (i.e. no conjugated). As a result, the entire molecule only has a spread of electrostatic
191 potential of 140.3 kJ mol⁻¹ compared to 377.8 kJ mol⁻¹ for terpinen-4-ol. This results in a
192 lack of specific sites for solvent interaction and thus it might be expected to have a poorer
193 solvation sphere in a more polar solvent.

194 Polarisability is the measure of how capable a compound is of inducing a dipole through its
195 molecular electron distribution. The correlation here is based more on the degree of
196 conjugation rather than functionality, so higher values were observed for sesquiterpenes
197 with the greater number of isoprene π -systems. Whilst this is considered a weaker effect
198 than the dipole moment with regards to solvent interactions, polarisability still helps define
199 preferential solvation in the solubilization of the investigated non-polar molecules in
200 aqueous media. Combinations of these factors contribute to governing the molecule-solvent
201 interactions, with the effects contributing to the formation of solvent shells and clustering.
202 These calculated values will be correlated with the real extraction performances to test
203 whether ESP, dipole moment and polarisability can predict extraction performance
204 dependent on the polarity of the still charge.

205 **Terpene extraction values**

206 The final distillate terpene concentrations of all distillations varied depending on the
207 compound (Table 4). The monoterpenes were most abundant with α -pinene showing the
208 highest concentrations, followed by β -myrcene and limonene. The one representative of the
209 oxygenated monoterpene fraction, terpinen-4-ol, ranged in concentration from 0.04 - 1.67
210 mg/L. The two sesquiterpenes α -humulene and β -caryophyllene showed a concentration
211 ranging from 0.07 - 1.84 mg/L and 0.31 - 3.29 mg/L respectively.

212 **Influence parameter**

213 *Botanical Ratio*

214 The results showed an increase in concentration of aroma compounds as the botanical ratio
215 is increased. Additionally, this parameter showed the greatest influence for all measured
216 terpenes using the Equation 1 to calculate the impact of all parameters (Figure 3).

217 Usually the content of essential oils is expressed in weight/weight percent and increasing
218 the amount of plant material increases the initial obtained essential oil content, as expected.
219 This parameter shows the greatest impact, although the extract is affected by the distillation
220 rate (19). It is worth noting however, that doubling the botanical ratio did not result in a
221 2-fold but rather a 1.5-fold increase in extraction for the compounds investigated.

222 *Charge amount*

223 The initial charge volume was influential on the final concentration of all terpenes studied.
224 Plots of the DOE (Figure 4) showed that increasing initial charge volume resulted in higher
225 extraction concentrations. The increased volume results in a prolonged distillation, and
226 therefore more time for the solvent to interact with the plant material, leading to a higher
227 extraction of terpenes as well as the higher collection volume. Existing EO extraction

228 research shows a similar trend of increased extraction when the distillation time was
229 extended (18). Further research suggests that an increase in distillation time resulted in
230 higher extraction values for mono- and sesquiterpenes due to the diffusive resistances in
231 subcutaneous glands within the plant material (40).

232

233 *Initial ethanol concentration*

234 The observed inverse trend was of the extracted concentration of terpinen-4-ol in relation to
235 the ethanol concentration of the charge was enhanced (Figure 5). Due to the difference in
236 dipole moment of this compound, as shown via the QSAR modelling, terpinen-4-ol being
237 polar and interacts more energetically with water. Jacotet-Navarro et al. (41) previously
238 determined an ideal theoretical solubility for ethanol water mixtures using COSMO-RS
239 comparing their calculations with experimental results. In their study they were able to find
240 a different ethanol concentration for rosmarinic and carnosic acid. Additional, studies on
241 vapour-liquid equilibrium of hydro-alcoholic binary solutions with aroma compounds, such
242 as terpenes at 101.3 kPa also revealed that the increase of the ethanol concentration led to a
243 decrease of the absolute volatility of the five terpenes investigated (42).

244 Furthermore, these measurements showed that oxygenated compounds are more volatile
245 than ethanol between 0 and 0.06 - 0.08 mole % ethanol. Results from the DOE also
246 revealed that this parameter had the least impact on other terpenes such as monoterpenes
247 and the sesquiterpenes. However, trace plots indicated a maximum between 50% ABV and
248 55% ABV ethanol optimum extraction of the monoterpenes, whereas the sesquiterpenes
249 showed a minimum in at 55% ABV ethanol concentration of the initial charge. Whilst
250 initial results suggest a decrease in extraction for terpinen-4-ol, and an increase for
251 limonene, β -myrcene and β -pinene as the ethanol concentration increased, some of the
252 other compounds are less clear. For the other compounds investigated, some display the
253 opposite change to that which was expected, and others give negligible deviation.

254 **Design of experiment model vs optimum condition.**

255 Confirmation experiments helped determine the variance between each observation and the
256 model results of the DOE, which showed an unequally distributed variance among each
257 terpene group (Figure 6). The monoterpenes α -pinene, γ -terpinene, β -pinene, showed the
258 smallest variance ranging from 5 to 15% whereas the greatest deviation was found on
259 β -caryophyllene, showing a variance range of 98%. Thus, the DOE equation is not
260 necessarily capable of describing all categories of terpenes. However, when considering
261 extractions of EO solely, the predicted value and the experimental value was in good
262 agreement (43, 44).

263

264 Sensory

265 Using sensory analysis to characterize each sample, the condition using 6 g/l, 400 mL, 45%
266 ABV was described as highest in piney, woody and citrus, herbaceous notes ($p < 0.05$)
267 which are conferred by α -pinene and γ -terpinene (45, 46). Furthermore, the condition using
268 2 g/L, 400 mL, 45% ABV was described the lowest in piney, woody, grapefruit, lime,
269 herbaceous, pepper and citrus, herbaceous notes ($p < 0.05$) correlating to the compounds
270 α -pinene, terpinen-4-ol and γ -terpinene. The sensory sample (6 g/L / 400 mL / 45%)
271 showed the third highest concentrations in α -pinene, and the highest concentration of
272 terpinen-4-ol and γ -terpinene of all tested distillates. However, using an untrained panel
273 which isn't screened or tuned reduces this effect on the sensory evaluation. It was possible
274 to correlate the concentrations of certain flavour compounds and the sensory results
275 revealing that a higher botanical ratio was shown to give higher values in the sensory
276 description, except for the samples using 65% ABV regarding the compound's limonene,
277 γ -terpinene and α -humulene. Similarly, a low botanical ratio led to lower flavour intensity
278 with most descriptors except for α -humulene and terpinen-4-ol (Table 5).

279

280

Conclusion

281 Three different distillation parameters were considered: botanical ratio, charge volume and
282 initial ethanol concentration in gin distillation via vapor infusion to understand their effect
283 on flavour and aroma compound extraction. A 3^3 factorial experimental design was
284 performed using small-scale laboratory distillation equipment with a vapor basket for
285 extraction. Density functional theory was used to model nine common flavour and aroma
286 compounds in juniper berries. This was computed to produce quantitative structure–activity
287 relationships including dipole moments, polarisability and electrostatic surface potential.
288 These calculations resulted in the highest values in case of the oxygenated monoterpene
289 terpinen-4-ol. This knowledge is of great use to understand the solvent interaction and the
290 behaviour in the still. The botanical ratio showed the highest influence based on the design
291 of experiment results followed by the initial charge volume determined by SPME-GC/MS.
292 These parameters can be used to alter the concentration of the terpenes, thus, influencing
293 the character of the final product.

294 Lastly, within the investigated concentration range the ethanol concentration of the charge
295 had the least influence on the extraction. Only in the case of terpinene-4-ol the ethanol
296 concentration had a noticeable influence showing that the extraction increases with lower
297 initial ethanol concentration. However, confirmation experiments of the DOE showed that
298 only in case of four terpenes (α -pinene, β -pinene, β -myrcene and α -humulene) was the
299 variance below 15%. Nevertheless, an untrained sensory panel assessed 6 specific
300 conditions and a correlation between the botanical ratio and the ethanol concentration was
301 noticeable. The present research shows possibilities how to influence and maximize the
302 extraction of certain flavour compounds as well as giving insight of these flavour
303 compounds and their chemometrics.

304

305

Author Contributions (CRediT)

306 Jan Hodel carried out experiments, data validation and writing; Michael Burke carried out
307 experiments, calculations and writing; Annie Hill was responsible for supervision, review
308 and editing.

309

310

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450

451 Table 1 Chemicals used for Gas Chromatography/ Mass Spectrometry and aroma
 452 characteristics

Compound	CAS	Group	Threshold values (46, 47)	Aroma
α -pinene	7785-70-8	Monoterpene	2.5 to 62	piney, woody ^(a)
β -pinene	18172-67-3	Monoterpene	140	woody, cooling ^(a)
α -phellandrene	99-83-2	Monoterpene	40 to 200	herbaceous, minty ^(a)
β -myrcene	123-35-3	Monoterpene	15	woody, rose, peach ^(b)
R-limonene	5989-27-5	Monoterpene	4 to 229	lemon, orange, sweet ^(a)
γ -terpinene	99-85-4	Monoterpene	1 mg/L	citrus, herbaceous ^(c)
terpinen-4-ol	2438-10-0	Oxygenated Monoterpene	130	grapefruit, lime, herbaceous, pepper ^(b)
β -caryophyllene	87-44-5	Sesquiterpene	64-90	spicy, woody ^(a)
α -humulene	6753-98-6	Sesquiterpene	0.23-63	balsamic, flowery, grassy, herbal ^(a)

453 Aroma characteristics from ^(a) (46), ^(b) (48), ^(c) (45) Threshold values in $\mu\text{g/L}$ if not indicated
 454 otherwise

455

456 Table 2 Factors of the design of experiment

Level	Factor A Botanical ratio [g/L]	Factor B Charge [mL]	Factor C EtOH concentration [ABV]
1	2 (-1)	200 (-1)	45 (-1)
2	4 (0)	300 (0)	55 (0)
3	6 (+1)	400 (+1)	65 (+1)

457

458 Table 3 Calculated QSAR properties for the investigated flavour compounds

Compound	Dipole Moment debye	Polarisability $\times 10^{-30} \text{ m}^3$	ESP	
			Min.	Max.
			kJ mol^{-1}	
α -pinene ⁽¹⁾	0.13	53.66	-83.53	51.85
β -pinene ⁽¹⁾	0.64	53.6	-96.1	56.66
β -myrcene ⁽¹⁾	0.56	55.08	-87.57	67.39
α -phellandrene ⁽¹⁾	0.14	54.47	-93.12	66.19
DL-limonene ⁽¹⁾	0.42	54.07	-90.38	57.39
γ -terpinene ⁽¹⁾	0.07	54.13	-88.7	51.63
terpinen-4-ol ⁽²⁾	1.23	54.93	-175.7	202.1
α -humulene ⁽³⁾	0.63	61.14	-105	58.17
β -caryophyllene ⁽³⁾	0.66	62.15	-93.75	58.55

459 ESP = electrostatic surface potential, ⁽¹⁾ = Monoterpene; ⁽²⁾ = Oxygenated Monoterpene; ⁽³⁾ =

460 Sesquiterpene

461

462 Table 4 Measurement values of DOE each condition from GC-MS

AB V [%]	Charge [mL]	Botanical ratio [g/L]	α -pinene	β -pinene	β -myrcene	α -phellandrene	limonene	γ -terpinene	terpinen-4-ol	β -caryophyllene	α -humulene
45	200	2	8.50	1.07	4.95	0.05	1.37	0.43	0.30	0.39	0.43
45	200	4	6.61	0.84	4.50	0.04	1.13	0.38	0.56	0.23	0.36
45	200	6	14.08	1.66	10.05	0.10	3.21	0.93	1.13	0.46	0.54
45	300	2	13.13	0.95	9.91	0.04	2.99	0.29	0.20	0.29	0.40
45	300	4	15.38	2.00	8.26	0.06	4.41	0.69	0.83	0.44	0.51
45	300	6	24.72	2.32	18.69	0.14	9.14	1.01	1.23	2.52	1.41
45	400	2	5.32	1.13	10.56	0.08	1.93	0.52	0.59	0.35	0.45
45	400	4	17.07	1.60	15.28	0.12	5.41	1.35	1.12	1.07	0.85
45	400	6	26.18	2.70	20.41	0.22	7.57	1.49	1.67	1.96	1.19
55	200	2	5.50	0.70	2.06	0.04	1.04	0.21	0.09	0.09	0.33
55	200	4	10.93	1.53	13.57	0.07	3.03	0.35	0.17	0.28	0.42
55	200	6	17.44	1.77	16.76	0.17	5.40	0.83	0.48	0.95	0.76
55	300	2	7.69	1.16	3.28	0.03	2.09	0.27	0.10	0.10	0.33
55	300	4	13.09	1.56	12.24	0.10	3.78	0.82	0.33	0.73	0.62
55	300	6	27.82	2.77	22.54	0.14	8.25	1.35	0.68	1.56	1.08
55	400	2	10.71	1.23	11.63	0.12	3.99	0.68	0.20	0.47	0.52
55	400	4	31.11	2.75	28.24	0.18	7.59	1.68	0.52	1.46	1.10
55	400	6	18.34	1.26	14.36	0.15	4.81	0.87	0.46	0.76	0.71
65	200	2	4.65	0.81	1.70	0.02	0.59	0.14	0.04	0.07	0.31
65	200	4	9.35	1.15	6.77	0.06	1.93	0.42	0.07	0.34	0.42
65	200	6	9.30	1.16	6.14	0.03	1.41	0.20	0.08	0.32	0.44
65	300	2	4.99	0.62	5.45	0.04	1.30	0.20	0.05	0.10	0.34

65	300	4	10.37	1.42	8.07	0.08	4.49	0.57	0.14	0.49	0.51
65	300	6	22.14	2.07	17.10	0.23	8.53	0.88	0.14	1.11	0.79
65	400	2	9.91	1.29	8.89	0.06	1.97	0.51	0.11	0.25	0.42
65	400	4	16.04	1.67	18.44	0.17	7.35	1.08	0.16	1.69	1.05
65	400	6	36.60	3.61	26.16	0.18	9.68	1.50	0.38	3.29	1.84

463 Concentrations in mg/L

464
465

Table 5 Chemicals used for Gas Chromatography/ Mass Spectrometry and odour characteristics

Compound	Descriptors	2 [g/L]	4 [g/L]/	6 [g/L]/	2 [g/L]/	4 [g/L]/	6 [g/L]/
		400 mL	400 mL	400 mL	400 mL	400 mL	400 mL
		45 % ABV			65 % ABV		
α -pinene	piney, woody	4.05	4.9	5.55	4.4	4.65	5.1
β -pinene	woody, cooling	4.55	4.55	4.6	5	4.65	5.1
α -phellandrene	herbaceous, minty	4.4	4.4	4.15	4.1	4.4	5
β -myrcene	woody, rose, peach	3.4	3.4	3.7	3.9	4.2	4.05
R-limonene	lemon, orange, sweet	3.15	4.15	4.55	3.7	3.85	3.5
γ -terpinene	citrus, herbaceous	3.5	4.45	5	3.8	4.6	4
terpinen-4-ol	grapefruit, lime, herbaceous, pepper	3.35	4.65	4.7	4.55	4.15	4.4
β -caryophyllene	spicy, woody	3.2	4.05	4.25	3.55	3.65	4.1
α -humulene	balsamic, flowery, grassy, herbal	3.95	4	4.2	4.25	4	4.15

466 Values are means of the quantitative descriptive analysis

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471 Reviewer comments:

472 Editor-in-Chief comments to the Authors

473 You have some constructive feedback from the reviewers particularly regarding the sensory data
474 and its robustness/value. I think the suggestion to expand the GCMS data and relegate the sensory
475 info is a good one. Pleasingly both reviewers are broadly supportive of your MS.

476 - please consult the instructions to authors -

477 <https://onlinelibrary.wiley.com/page/journal/20500416/homepage/forauthors.html> - as the journal
478 names in the references need to be in their abbreviated form (and also not in capitals).

479 - as requested previously label the constituent parts of the distillation apparatus (perhaps a better
480 description than 'set up').

481 - The Figures need tidying up. its mL not ml - polarizability and standardized, replace the z with an s,
482 titles remove 'simplified', 'all the confirmations runs' could be better phrased, Fig 4 what are the units
483 of the y axis, fig 6 45 % to 45% etc etc, Fig 2 the colour histograms are li) too small and 9ii0
484 inconsistently sized, m3 as L

485

486 Reviewer (1) comments to the Authors

487 This is an interesting paper that examines how the key flavour components of juniper distil under
488 different conditions. This information is useful for gin distillers. The chemistry appears to be sound,
489 though there are a couple of points to note:

490 • Line 109: how were the nine key flavour compounds selected? - give a reference

491 • It would be useful to present the full results of the GC-MS analysis in a table

492 Although the analytical aspects of this paper look sound, the sensory work is questionable. The
493 sensory work is not described in the abstract, which indicates that it is not a critical part of the study.
494 Also, the approach used is not a standard one. Typically, a sensory panel would be asked to rate the
495 intensity of key flavour attributes, such as piney, citrus, woody, with the results then compared to the
496 compositional data. In this study, the panel have been trained to score the intensity of each of the
497 flavour compounds, having first been trained with standard samples of these. There is little point in
498 doing this when you already have analytical data showing the levels of these compounds. This
499 analytical data will be much less noisy and more reproducible than data from a sensory panel. There
500 is also a lot of overlap in sensory terms between the compounds. For example, four of the nine
501 compounds are described as having woody notes, three as having citrus notes, etc. So, I would
502 question whether the panellists could reliably discriminate one compound from another. There is no
503 data to confirm that this has been evaluated. Also, a group of twenty panellists is not enough for a
504 preference test. This group would not be representative of consumers, and the authors do not
505 indicate if these individuals are regular gin drinkers.

506 Before this paper can be accepted from publication, the authors either to justify the sensory
507 approach used or repeat the analysis using a standard method. The other option is to consider
508 whether the sensory data is a critical part of this paper. If not, it could potentially be removed.

509

510 Reviewer (2) comments to the Authors

511 I cannot understand why you did not discuss your own previous paper on the subject (See Ref 34) in
512 the background discussion. In my opinion it is the most relevant and interesting and puts this
513 current paper into context, for instance I could not understand why you chose to use vapour infusion

514 as a first choice until I read Ref 34 which was hidden in the experimental. As soon as I read this all
515 became clear.
516 The use of DOE was good.
517 I liked the attempts you made to theoretically predict extraction efficiencies of particular botanicals
518 using QSAR and ESP although. Personally I would make use of CalcLogP software that medicinal
519 chemists use which is probably the most relevant software because you can then compare it to
520 actual logP values from measuring solubilities in octanol/water but that does not detract from a very
521 interesting paper.
522