Comparison of essential oils of clove buds extracted with supercritical carbon dioxide and other three traditional extraction methods

Guan Wenqiang a, Li Shufen a,*, Yan Ruixiang b, Tang Shaokun a, Quan Can a

a Key Laboratory for Green Chemical Technology of State Education Ministry, School of Chemical Engineering and Technology, Tianjin University, Tianjin 300072, PR China

b National Engineering and Technology Research Center for Preservation of Agricultural Products, Tianjin 300384, PR China

Received 19 December 2005; received in revised form 3 March 2006; accepted 6 April 2006

Abstract

Supercritical fluid extraction (SFE) of essential oil from clove buds with CO₂ was explored. The effect of different parameters, such as temperature (30 °C, 40 °C, 50 °C), pressure (10 MPa, 20 MPa, 30 MPa) and particle size (three degree index), on the extraction yield and the content of eugenol in extracts was investigated using three-level orthogonal array design. The experimental results show that the temperature has the largest effect on the eugenol content of the extracts, and particle size has the maximum effect on the oil yield. The essential oil of 19.56% yield, in which the maximum content of eugenol in extracts is 58.77%, can be extracted from clove buds at pressure of 10 MPa and temperature of 50 °C. Essential oil of clove buds obtained by SFE, hydrodistillation, steam distillation and Soxhlet extraction were further analyzed by gas chromatography/mass spectrometric detection to compare the extraction methods. Twenty three compounds in the clove oils have been identified, showing that the composition of the clove oil extracted by different methods is mostly similar, whereas relative concentration of the identified compounds is apparently different. General characteristics of the clove oils obtained by different methods were further compared, and SFE is considered as the optimum process among the four processes for obtaining clove oil with high quality.

Keywords: Supercritical fluid extraction; Carbon dioxide; Essential oil; Clove bud; Steam distillation; Hydrodistillation; Soxhlet extraction

1. Introduction

Clove (Eugenia caryophyllata Thunb.) is widely cultivated in Madagascar, Sri Lanka, Indonesia and the south of China (Bureau of Drug Administration of China, 1989). Clove bud oils have biological activities, such as antibacterial, antifungal, insecticidal and antioxidant properties, and are used traditionally as flavoring agent and antimicrobial material in food (Huang, Ho, Lee, & Yap, 2002; Lee & Shimamoto, 2001; Velluti, Sanchis, Ramos, & Marin, 2003). For example, clove oil was effective against L. monocytogenes and S. Enteritidis in tryptone soya broth (TSB) and cheese (Smith-Palmer, Stewart, & Fyfe, 1998, 2001). The high levels of eugenol contained in clove essential oil give it strong biological activity and antimicrobial activity. This phenolic compound can denature proteins and reacts with cell membrane phospholipids changing their permeability (Briozzo, 1989; Deans & Ritchie, 1987). Clove oil also has several therapeutic effects, including antiphlogistic, antivirus, analgesic, antispasmodic, antiruminative, kidney reinforcement, antiseptic, HCMV extracorporeal restraining effect (Liu, Mao, & Hong, 1997; National Pharmacopoeia Committee of China, 2000). In Korea, clove oil has been successfully used for asthma and various allergic disorders by oral administration (Kim, Lee, & Hong, 1998).

The essential oil of aromatic herbs is traditionally obtained by hydrodistillation, steam distillation, or solvent
2. Materials and methods

2.1. Materials

The buds of clove (E. caryophyllata Thunb.) were purchased from Tianjin factory for Chinese herbs. The samples were dried at 30 °C in a ventilated drying oven and stored in plastic bags at ambient temperature and protected from light. The samples were ground by a FW80 Sample Mill (Taisite CO., Tianjin, China) in different particle sizes to get the different particle distribution, which was measured by mechanical sieving after extraction and calculated by weight of different size of clove bud particle. Grades of particle size was classified on the following scale: 1 > 10 mesh; 2 = 10–20 mesh; 3 = 20–40 mesh; 4 = 40–60 mesh; 5 = 60–80 mesh; 6 = 80–100 mesh; 7 = 100–120 mesh; 8 < 120 mesh. Particle size index was calculated by following formula: Particle size index = \( \sum (\text{weight of each grade} \times \text{grade})/(\text{total weight} \times \text{highest grade}) \). The particle size index of material in this experiment was 0.7944, 0.6430, 0.5223, and named as 1#, 2# and 3#, respectively.

The solvents and chemicals were obtained from following sources: carbon dioxide, 99.99% purity, from Tianjin Anxing gas factory; China. Dichloromethane, purity > 99.9%, from Tianjin chemical Co., China; Eugenol, Purity ≥ 99.0% (GC), from Fluka Co., Germany; Eugenol acetate, Purity > 90.0% (GC), TCI America Co., Japan.

2.2. Experimental apparatus and methods

2.2.1. Supercritical CO2 extraction

Extraction of essential oil from clove buds was experimentally determined using the Speed SFE instrument (Applied Separations Inc., Allenton, PA, USA). Liquid CO2 was pressurized with a high-pressure pump and then charged into the extraction column to desired pressure. The pressure was controlled to an accuracy of about 1% over the measuring range. The extraction column was 32 ml with 14.40 mm inner diameter and 195 mm length, being packed with powdered raw materials and glass beads. The extraction column was heated with an oven and its temperature was indicated and controlled by a thermocouple to within ±1 °C. The supercritical CO2 with dissolved compounds passed through a heated micrometer valve, and was subsequently expanded to ambient pressure. The extract was precipitated in a collect vial at ambient pressure and temperature. A calibrated wet-test meter at known temperature and pressure measured the total amount of CO2.

For each extraction test, the extractor was charged with about 15 g of ground clove bud powder. CO2 flow rates ranging about 2 l/min were used. The oil weight was measured by precision balance until no oil was extracted out from the clove bud powder.

2.2.2. Hydro and steam distillation

The plant (100 g of dried and ground clove buds) in 500 ml flask was submitted to hydrodistillation for 4–6 h and steam distillation for 8–10 h. The volatile distillate was collected until no oil drop out. The distillate was saturated with sodium chloride and added with some ether. Then, the ether layer and hydro layer were separated by funnel. After dehydrated by anhydrous sodium sulphate, the ether layer was further heated in 60 °C water bath to make oil to be concentrated and the ether to be recovered. The oil was weighed and refrigerated prior to analysis.
2.2.3. Solvent extraction

The ground clove bud samples (30 g) were weighed and quantitatively transferred into a filter paper extraction thimble and inserted into a 500-ml reflux flask, then extracted with 250 ml absolute ethanol for about 6 h in a Soxhlet apparatus (Quan et al., 2004). After Soxhlet extraction, extracts were concentrated using rotary vacuum evaporator at 50 °C.

2.2.4. GC and GC/MS analysis

GC analyses were performed using a Shimadzu GC-2010 gas chromatograph equipped with a FID and a DB-5 fused-silica column (30 m × 0.25 mm i.d., film thickness 0.25 μm, Agilent). Oven temperature was 80 °C for 2 min, then programmed heating from 80 to 230 °C at a rate of 6 °C/min, and at 230 °C for 2 min. Injector and detector temperatures were 230 °C. The carrier gas, nitrogen, was adjusted to a linear velocity of 24 ml/min. The samples (0.1% in absolute ethanol) were injected into the GC by split mode with a split ratio of 1/20. Methyl salicylate was used as internal standard.

GC/MS analyses were performed using Finnigan Trace GC/DSQ equipped with a BTX.5ms fused-silica capillary column (15 m × 0.25 mm i.d., film thickness 0.25 μm, RESTEK CORP.). Oven temperature conditions were: 40 °C for 1 min, then programmed heating from 40 to 200 °C at a rate of 6 °C/min and from 200 to 280 °C at a rate of 30 °C/min. Injector temperature was 250 °C. The carrier gas, helium, was adjusted to a linear velocity of 1 ml/min. Ion source temperature was 250 °C; The ionization energy was 70 eV with a scan time of 1 s and mass range of 20–500 AMU. Samples were run in dichloromethane with a dilution of 0.1% (V/V). Compounds were identified by matching their mass spectra and retention times with those of pure compounds whenever possible. NIST (National Institute of Standards and Technologies) Mass Spectra Library was also used as a reference (see Table 1).

3. Results and discussion

3.1. Yield and eugenol content of the oil extracted by SFE

The following parameters was used: temperature, 30 °C, 40 °C and 50 °C; Pressure, 10 MPa, 20 MPa and 30 MPa; particle size, 1#(0.7944), 2#(0.6430) and 3#(0.5223). All the selected factors were examined by using a three-level orthogonal array design with an OA9 (3³) matrix. Table 2 shows the experimental conditions for each of the SFE runs. It can be seen from the order of the maximum differences that the particle size had the most influence on the oil yield, then the temperature and pressure. However, the sequence of the influences of the parameters on the eugenol content in the oils was temperature, particle size, and pressure. That is, the factor of temperature has the maximum influence on the eugenol content in the oils.

Fig. 1 shows the effect of temperature, pressure and particle size on the yield and eugenol content of clove

Table 1
Three-level orthogonal design and experimental results for extraction of clove oil with SC–CO2

<table>
<thead>
<tr>
<th>Run No.</th>
<th>Factor A (T/°C)</th>
<th>Factor B (P/MPa)</th>
<th>Factor C (particle size/#)</th>
<th>Yield (kg extract/kg feed)</th>
<th>Eugenol content (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1(30)</td>
<td>1(10)</td>
<td>1(1#)</td>
<td>0.2056</td>
<td>53.69</td>
</tr>
<tr>
<td>2</td>
<td>1(30)</td>
<td>2(20)</td>
<td>2(2#)</td>
<td>0.1943</td>
<td>54.22</td>
</tr>
<tr>
<td>3</td>
<td>1(30)</td>
<td>3(30)</td>
<td>3(3#)</td>
<td>0.1830</td>
<td>55.64</td>
</tr>
<tr>
<td>4</td>
<td>2(40)</td>
<td>1(10)</td>
<td>3(3#)</td>
<td>0.1910</td>
<td>56.20</td>
</tr>
<tr>
<td>5</td>
<td>2(40)</td>
<td>2(20)</td>
<td>1(1#)</td>
<td>0.2224</td>
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</tr>
<tr>
<td>6</td>
<td>2(40)</td>
<td>3(30)</td>
<td>2(2#)</td>
<td>0.2043</td>
<td>55.30</td>
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<td>7</td>
<td>3(50)</td>
<td>1(10)</td>
<td>2(2#)</td>
<td>0.1956</td>
<td>58.77</td>
</tr>
<tr>
<td>8</td>
<td>3(50)</td>
<td>2(20)</td>
<td>3(3#)</td>
<td>0.1827</td>
<td>57.83</td>
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<tr>
<td>9</td>
<td>3(50)</td>
<td>3(30)</td>
<td>1(1#)</td>
<td>0.2395</td>
<td>56.97</td>
</tr>
</tbody>
</table>

Yield (%)

\[
\text{K1} = 0.5830
\]
\[
\text{K2} = 0.6178
\]
\[
\text{K3} = 0.6179
\]
\[
\text{K1/3} = 0.1943
\]
\[
\text{K2/3} = 0.2059
\]
\[
\text{K3/3} = 0.2060
\]
\[
\text{R} = 0.0117
\]

\[
\sum = 1.8186
\]

Eugenol content (%)

\[
\text{K1} = 163.55
\]
\[
\text{K2} = 166.02
\]
\[
\text{K3} = 173.57
\]
\[
\text{K1/3} = 54.52
\]
\[
\text{K2/3} = 55.34
\]
\[
\text{K3/3} = 57.86
\]
\[
\text{R} = 3.34
\]

\[
\sum = 503.14
\]
<table>
<thead>
<tr>
<th>No.</th>
<th>Compound</th>
<th>RT&lt;sup&gt;a&lt;/sup&gt;</th>
<th>Run 1</th>
<th>Run 2</th>
<th>Run 3</th>
<th>Run 4</th>
<th>Run 5</th>
<th>Run 6</th>
<th>Run 7</th>
<th>Run 8</th>
<th>Run 9</th>
<th>SD&lt;sup&gt;b&lt;/sup&gt;</th>
<th>HD&lt;sup&gt;c&lt;/sup&gt;</th>
<th>SO&lt;sup&gt;d&lt;/sup&gt;</th>
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<td>0.46</td>
<td>0.45</td>
<td>0.44</td>
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<td>Eugenol</td>
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<td>54.58</td>
<td>55.07</td>
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<td>55.14</td>
<td>57.36</td>
<td>56.81</td>
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<td>48.82</td>
<td>57.24</td>
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<td>α-Copaene</td>
<td>13.34</td>
<td>0.95</td>
<td>0.63</td>
<td>0.86</td>
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<td>2.15</td>
<td>2.13</td>
<td>2.02</td>
<td>1.9</td>
<td>2.08</td>
<td>1.95</td>
<td>2.61</td>
<td>4.41</td>
<td>2.03</td>
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<td>0.28</td>
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<td>0.22</td>
<td>0.22</td>
<td>0.22</td>
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<td>0.22</td>
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<td>0.16</td>
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<tr>
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<td>α-Farnesene</td>
<td>16.1</td>
<td>0.25</td>
<td>0.23</td>
<td>0.29</td>
<td>0.32</td>
<td>0.22</td>
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<td>0.2</td>
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<td>Unidentified</td>
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<td>0.07</td>
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<td>1.05</td>
<td>1.46</td>
<td>0.72</td>
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<td>Cubenol</td>
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<td>0.41</td>
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<td>Caryophyllene oxide</td>
<td>17.41</td>
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<td>Diethyl Phthalate</td>
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<td>0.35</td>
<td>0.33</td>
<td>1.1</td>
<td>0.74</td>
<td>1.43</td>
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<tr>
<td>18</td>
<td>1,4-Methanoazulen-7(1H)-one, octahydro-4,8-</td>
<td>17.92</td>
<td>0.11</td>
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<td>–</td>
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<td>19</td>
<td>Tetracyclo[6.3.2.0(2,5).0(1,8)]ridecan-9-ol, 4,4-dimethyl-</td>
<td>18.35</td>
<td>0.11</td>
<td>0.14</td>
<td>0.13</td>
<td>–</td>
<td>0.14</td>
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<td>Tetracyclo[6.3.2.0(2,5).0(1,8)]ridecan-9-ol, 4,4-dimethyl-</td>
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<td>Isoaromadendrene epoxide</td>
<td>19.07</td>
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<td>0.12</td>
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<td>0.25</td>
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<td>0.28</td>
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<tr>
<td>22</td>
<td>2,2,4-Trimethyl-3-(3,8,12,16)-tetramethyl-heptadeca-3,7</td>
<td>30.6</td>
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<td>0.24</td>
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<tr>
<td>23</td>
<td>Tetratetracontane</td>
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<td>–</td>
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<td>–</td>
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<td>0.22</td>
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<td>–</td>
<td>–</td>
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</tr>
</tbody>
</table>

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<sup>a</sup> Retention time on BTX 5ms fused silica capillary column.  
<sup>b</sup> Steam distillation.  
<sup>c</sup> Hydrodistillation.  
<sup>d</sup> Soxhlet extraction.
oil extracted by supercritical CO₂. It can be observed that
the increase of temperature from 30 °C to 40 °C results in
the increase of the extraction yield and high eugenol con-
tent in the oils, while the increase of temperature from
40 °C to 50 °C does not result in the increase of the oil
yield, and there is increase of eugenol content in clove
oil. This is ascribed to the decrease of the CO₂ density
with increasing temperature, which dominates over the
increase of the solute vapor pressure at this certain pres-
sure (Lou, Folas, Voutasa, & Magoulas, 2003). As it
was expected, the extraction yield enhanced significantly
with increase of pressure, due to the increase of the solu-
bility of the oil components. This is attributed to the
increase of the CO₂ density, which results in the increase
of its dissolving ability. However, as the high-molecular
weight compounds in clove buds (fatty acids, fatty acids
methyl esters, sterols, etc.) were also co-extracted with
increase of the pressure, the eugenol content of the clove
oil does not change obviously. Therefore, the highest
eugenol content of the clove oil is extracted at 50 °C
and 10 MPa.

The extraction yield increases by decreasing the particle
size of the comminuted clove buds. It is due to the higher
amount of oil released as the bud cells are destroyed by
milling (Mostafa et al., 2004; Reverchon, 1997), and this
amount of oil is easily extracted for direct exposure to
the supercritical CO₂. Moreover, shorter diffusion paths
in the milled solid matrix result in a smaller intraparticle
resistance to diffusion. However, the eugenol content in
the clove oil increases as the particle size increase. There-
fore, the particle size should not be too small if the aim
is to extract the volatile ingredients and to avoid more com-
 pounds with high-weight molecules to be co-extracted. It is
better for the particle size index to be controlled at about
0.6430(2#).

3.2. Composition of clove oil obtained by different methods

Table 2 lists the composition of clove oil obtained by dif-
ferent methods according to the results of GC–MS. It can
be seen that, twenty three compounds in the clove oils have
been identified, in which eugenol, β-caryophyllene and
eugenol acetate are the main components of clove oil. The
composition of the clove oil extracted by different methods
is mostly similar, whereas relative concentration of the iden-
tified compounds is apparently different. Clove oil obtained
by steam distillation contained highest percentage of euge-
nol (58.2%), then did oil obtained by SFE (53.8–55.9%)
Clove oil obtained by hydrodistillation had the lowest per-
centage of eugenol (48.82%). However, clove oil obtained
by SFE contained the highest percentage of eugenol acetate
(20.32–21.75%), which is also the main antioxidant ingredi-
ents in clove oil (Lee & Shibamoto, 2001). The highest per-
centage of active antioxidant ingredients, eugenol together
with eugenol acetate, in the extracted clove oil by SFE
was therefore obtained. The content of eugenol and eugenol
acetate in the extracted clove oil by Soxhlet method was also
higher than those by the two kinds of distillation methods.
Thermal degradation of eugenol acetate may take place
during hydro and steam distillation as eugenol acetate
was only 13.84%, 3.89%, respectively.

Besides eugenol, and eugenol acetate in clove oil, rela-
tive content of β-caryophyllene in the clove oil extracted
by steam distillation, hydrodistillation, SFE and Soxhlet
method was as high as 20.52%, 36.94%, 13.99–17.77%
and 17.5%, respectively. It was reported that β-caryophyl-
lene showed anti-inflammatory activity in several experi-
ments (Ghelardini, Galeotti, Di Cesare Mannelli, Mazzanti, & Bartolini, 2001). If the aim was to obtain high
content of β-caryophyllene in the clove oil, it maybe neces-
sary to choose hydrodistillation.
It can also be seen that the oil obtained by SFE contains small amount of co-extracted cuticular waxes. This result is similar to other author’s work (Mostafa et al., 2004; Myint et al., 1996).

3.3. Comprehensive comparison of the clove oils obtained by different methods

Color and texture are the prime characteristics and quality factors of essential oil, and extraction yield and extraction time are the important factor for the industrialization. Therefore, comprehensive comparisons of the clove oils obtained by different methods were further listed in Table 3. In Table 3, the content of eugenol and eugenol acetate was determined by GC.

It can be seen from Table 3 that, the content of the main biological ingredients of eugenol plus eugenol acetate in the clove oil by Soxhlet extraction is the lowest although its yield of the clove oils is the highest among the four extraction methods. Furthermore, the extracts by Soxhlet method is brown ointment, which means more undesired impurities and organic solvent residue may existed. SFE offers the most important advantages over other methods. Extraction yield of SFE was about two times as high as that obtained by steam and hydrodistillation. The highest content of eugenol plus eugenol acetate in the extracted oil was obtained. Pale yellow oil is desired and shortest extraction time is needed for SFE compared with other three extraction methods. Additionally, using supercritical CO2 instead of some harmful organic solvents would result in “greener” processes.

4. Conclusions

In the investigation of the effect of the three tested parameters (pressure, temperature and particle size) on the SFE extraction of essential oils from clove bud within the experimental domain. Particle size had the most apparent effect on the yield of essential oil; temperature was the most important factor for determining the content of eugenol of the clove oil. The possibility of manipulating the eugenol content of the clove bud oil by changing the parameters of the extraction is attainable in SFE.

The composition and some character of clove oil obtained by SFE and other methods were compared. Although compositions of the oils obtained by SFE and steam, hydrodistillation are similar, they do differ quantitatively. Extraction yield of SFE was about two times as high as that obtained by steam and hydrodistillation. SFE offers many important advantages over other three traditional methods, including higher extraction yield, the highest percentage of active antioxidant ingredients of eugenol together with eugenol acetate in the extracted clove, shorter extraction time, and so on. Therefore, SFE is considered as the optimum process for obtaining clove oil with high quality.

Acknowledgements

The authors are grateful for the financial support received from Tianjin Academy of Agricultural sciences through project PRESIDENTFUND-2004018, which made this study possible, and give a special thanks to Gao Xiaochen (School of Chemical Engineering and Technology, Tianjin University) for his collaboration and participation in this study.

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