

Essential Oil from Ether-Containing Plants of Juniper (*Juniperus*) and Spruce (*Picea*) Leaves by Distillation

Baktygul Abylaeva¹, Tilebaldy Abdulazizov², Zamira Sandybaeva¹, Zhyrgagul Abdullaeva^{3,4*} ,
Ernis Bepiev¹, Zakhro Akhmedova⁵

¹Departemnt of Pharmacy and Technology of Medications, Osh State University, Osh, Kyrgyzstan

²Department of General Chemistry and Ecology, Osh State University, Osh, Kyrgyzstan

³International Medical Faculty, Jolon Mamytov Campus, Osh State University, Osh, Kyrgyzstan

⁴Science and Research Department, Osh State University, Osh, Kyrgyzstan

⁵Institute of Microbiology, Academy of Sciences of the Uzbek Republic, Tashkent, Uzbekistan

Email: *jypar.science@oshsu.kg

How to cite this paper: Abylaeva, B., Abdulazizov, T., Sandybaeva, Z., Abdullaeva, Z., Bepiev, E. and Akhmedova, Z. (2020) Essential Oil from Ether-Containing Plants of Juniper (*Juniperus*) and Spruce (*Picea*) Leaves by Distillation. *Green and Sustainable Chemistry*, 10, 109-116.
<https://doi.org/10.4236/gsc.2020.104008>

Received: August 26, 2020

Accepted: October 18, 2020

Published: October 21, 2020

Copyright © 2020 by author(s) and Scientific Research Publishing Inc.

This work is licensed under the Creative Commons Attribution International License (CC BY 4.0).

<http://creativecommons.org/licenses/by/4.0/>



Open Access

Abstract

This article is investigating the preparation of essential oil from the ether-containing plants of Juniper (*Juniperus*) and spruce (*Picea*) leaves by using the simple water distillation equipment. This water distillation method forms an essential oil with water vapor from plants of Juniper (*Juniperus*) and spruce (*Picea*) leaves based on Dalton's law of partial pressure, applicable for plants containing a large amount of ether oil where distillation temperature is not affecting product quality. Obtained essential oils are widely used in medicine, perfumery, cosmetics, and food industry. In addition, these essential oils possess different activities including antimicrobial, antiviral, and anti-inflammatory, which have different effects such as adaptogenic, antidepressive, disinfection, and wound healing, diuretic, and antipyretic.

Keywords

Essential Oil, Extract, Juniper, Spruce, Leaves, Distillation, Yield

1. Introduction

Ether containing trees *Juniperus* and *Picea* are widely distributed indigenous species in the Southern region of Kyrgyzstan (**Figure 1**). The kind *Juniperus* belongs to the *Cupressaceae* family, which is characterized by a large amount of essential oil in needles, wood, and seeds [1]. Essential oil components represented in **Table 1** and chemical constituents obtained from the *Juniperus* trees are including α -pinene, β -pinene, apigenin, sabinene, β -sitosterol, campesterol, limonene



Figure 1. (a) Spruce (*Picea*) leaves and (b) juniper (*Juniperus*) leaves.

Table 1. Essential oil components obtained from the *J. communis L.* [2].

	Rt	%
Monoterpene hydrocarbons		
1) α -Pinene	4.40	1.95
2) dl-Limonene	6.33	0.96
3) α -Pinene	10.78	0.80
4) (+)-4-Carene	12.44	3.86
5) Bicyclo[4.1.0]hept-2-ene,3,7,7-trimethyl	12.81	0.71
Sesquiterpene hydrocarbons		
1) α -Cedrene	12.98	0.15
2) α -Cadinene-4,9-diene	13.08	0.93
3) Cedrene	13.64	4.04
4) Gamma. 1-cadinene	14.09	1.00
Oxygenated monoterpenes		
1) 1-Indanone	7.60	1.15
2) Linalool	7.85	2.34
3) 2,3,3-Trimethyl-3-cyclopentene acetaldehyde	8.35	2.09
4) 5-Decene-1-ol	10.89	2.60
Oxygenated sesquiterpenes		
1) Cedrene epoxide	18.94	2.79

and many other compounds [2]. Other applications of the *Juniperus* reported as a biomonitor for heavy metal pollution due to mineral nutrients and excessive metal (Ca, Cd, Cr, Cu, Fe, K, Mg, Mn, Na, Pb, and Zn) accumulation effects [3].

Picea known as a spruce tree, belongs to the family of *Pinaceae* [4]. The chemical composition of volatile oils isolated from different species depends on geographic origin [5]. Antibacterial [6], anti-inflammatory [7], antifungal activity [8], analgesic activity [9], antidiabetic and antihyperlipidemic, antimicrobial activities [10] [11], antioxidant activity [12], antihypercholesterolemic activity

[13] and neuroprotective activity in Parkinson's disease [14] of *Picea* tree were studied. *Juniperus* species are used for the treatment of hyperglycemia, tuberculosis, bronchitis, pneumonia, ulcers, intestinal worms, to heal wounds and cure liver disease; berries are used in the treatment of skin diseases including skin rash and eczema, respiratory tract diseases, asthma, common cold, cough, bronchitis, throat inflammation, pneumonia and tuberculosis [11] [15]. In this regard, the study of the production and use of essential oil from ether-containing plants, including the leaves of spruce and juniper leaves is relevant.

Essential oil preparation from the juniper (*Juniperus*) and spruce (*Picea*) leaves by continuous water distillation and other methods studied [16] [17]. The antimicrobial activity of essential oils is attributed to a number of terpenoid and phenolic compounds in plants studied [18]. The essential oil of three species was analyzed for chemistry and antimicrobial activity [19].

The purpose of this work was to obtain essential oils and oil extracts from the local ether containing plants of juniper (*Juniperus*) and spruce (*Picea*). One of the approaches to solve these issues is the installation of laboratory tools and further development of technological equipment followed by identification of influence on technological factors, yield, and composition of essential oils.

2. Materials and Methods

Preparation of Raw Materials (Collection of Essential Oil Plants, Drying)

The essential oil is extracted directly from the fresh leaves. In the laboratory, the Soxhlet apparatus used to extract the essential oils from plants. Collected leaves of both species cleaned and stored in a refrigerator, in plastic bags at a temperature of 2°C - 4°C. Leaves of juniper and spruce harvested in the undergrowth, in mixed birch forests (age 10 - 15 years). Humidity was determined by drying the sample in an oven at 105°C ± 4°C in duplicate. The method of distillation of essential oil with water from plant materials is based on properties such as volatility and insolubility in water. The objective of the research was to isolate the essential oil in the initial stage of its distillation at different temperatures of the working steam.

The experiments were carried out under laboratory conditions in the temperature range of 100°C - 200°C. The yield of essential oils was determined by titration, the component composition studied by chromatography. When conducting experiments with both juniper leaves and spruce leaves, the process temperature significantly affects the oil yield.

Further, for the distillation of the essential oil, the plant material was extracted with water and carried out with a distillation apparatus in a water bath at a temperature of 178°C - 180°C. Water vapor from the steam generator passes through the plant material and volatile essential oil condensed into the refrigerator and then collected in the receiver. The essential oil of juniper and spruce leaves isolated and investigated in laboratory conditions as shown in **Figure 2**.

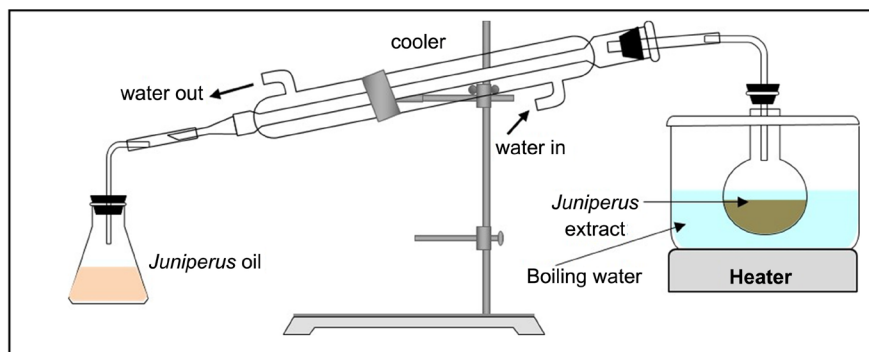


Figure 2. Experimental equipment for obtaining essential oils from juniper and spruce extracts by distillation.

3. Results and Discussions

The results of the distillation of the essential oil from the leaves of juniper and spruce with water vapor at different temperatures are shown in **Table 2**. The yield of the essential oil was determined by the titration method, and the component composition was studied by chromatographic methods. When conducting experiments with both juniper leaves and spruce leaves, the process temperature significantly affects the oil yield.

As the table shows, the yield of essential oil depends on the type of raw material obtained and on the temperature of the working steam. The data obtained compared with these indicators with its exhaustive distillation. With increasing temperature, the oil yield decreases in this case (**Figure 3**) other components begin to caramelize *i.e.* a sharp increase in the yield of monoterpenes observed [20].

Distillation of essential oils was carried out both from fresh and from dried materials. At the temperature from 100°C to 130°C, the yield increased with a slight excess in case of juniper leaves. According to [21] [22], the composition of essential oils contains aromatic compounds, terpenes, and terpenoids, aldehydes, saturated and unsaturated hydrocarbons, organic acids and amines, as well as heterocyclic compounds, organic sulfides, etc. With increasing in temperature (at 180°C and 200°C), the oil yield decreases, this is explained by an increase in the oxidative and polycondensation transformations of terpenoids, their conversion to resinous products [23]. The oil from spruce leaves is 1.8% at temperature 178°C, and for juniper leaves oil obtained from 2.1% at 187°C. The rate of release of essential oil from raw materials gradually increases with increasing temperature of the working steam *i.e.* the oil yield starts from 150 - 187°C.

According to the experimental data, a sufficiently complete distillation of essential oil is carried out at 100°C for 180 min, 130°C - 120 min, 150°C - 100 - 110 and 180°C - 70 - 80 min. This dependence in the initial stage has the same parameters and is determined by the temperature of the vapor. As the results of analyses show, the temperature for the release of essential oil from the raw materials used by in this work was 178°C - 186°C, and with the change in temperature, a change in the rate of oil release was observed.

Table 2. The distillation of the essential oil from the leaves of juniper by water vaporization at different temperatures.

Distillation temperature, °C	<i>Juniperus</i> leaves 100 g/ml	<i>Picea</i> leaves 100 g/ml
	Oil yield, %	Oil yield, %
100	-	-
130	0.06	0.08
150	0.8	1.6
178	1.8	1.8
187	2.1	-

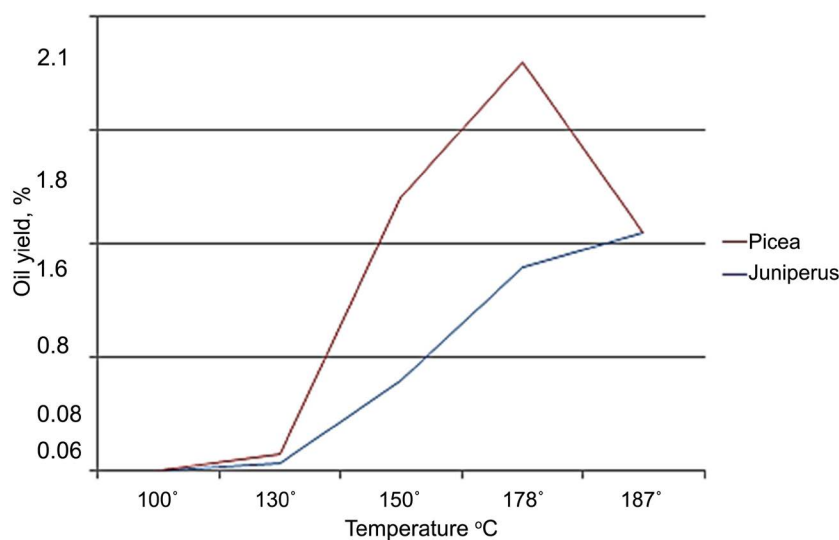


Figure 3. The dynamics of the allocation of essential oil at different temperatures of the working steam.

Based on experimental studies, the isolation of essential oil from juniper leaves is 74 minutes at a temperature of 186°C - 187°C, and from spruce leaves - 88 minutes at 178°C. With steam distillation, it takes at least 5 - 6 minutes at a temperature of 170°C - 180°C to isolate the essential oil. The results obtained in **Figure 3** indicate that amount of essential oil released increases when juniper leaves are used as a raw material in the processing of harvested leaves from the second half of August to the end of October.

The conducted studies allowed us to obtain data on potential reserves of terpenoids, varying their component composition, which allows us to further optimize the quantity and quality of marketable products. At the same time, the amount of oil in the woody greens of trees of different ages, both taking into account and without taking into account the main biocenotic factors, is almost the same, which is a positive point, because a sufficient yield of essential oil from averaged raw materials provided.

The essential oil extraction process is exponential. At the initial stage, intensive extraction of terpenoids characterized and a sharp decrease in their quantity



Figure 4. (a) Extracts of juniper and spruce; (b) essential oils of juniper and spruce obtained by distillation.

at the end of distillation observed. With the same dynamics of oil evolution at different temperatures of the working steam, it yields significant changes. The maximum amount of terpenoids is distilled off at 170 °C - 180 °C, those. Lowering and raising the process temperature leads to a decrease in oil volume. In the first case, incomplete selection occurs. With a further increase in temperature, the oxidative and condensation processes of the components of the essential oil are intensified. Juniper and spruce extracts, and essential oils obtained by distillation are shown in **Figure 4**. The relationship between the yield of essential oil and the distillation temperature expressed by the equation:

$$Y_1 = 2.1 + 187I + 128t$$

$$Y_2 = 1.8 + 178I + 86t$$

here, Y is the essential oil yield (%), I is the distillation temperature (°C), and t is the duration of distillation (min) (**Figure 4**).

When releasing the essential oil, a sufficiently significant factor is also the flow rate of the working steam. Increase and decrease in temperature reduces the yield of volatile terpenoids by 8% - 12%, due to incomplete distillation or losses in partial decomposition of the oil components.

4. Conclusions

Grinding the raw material to a size of 5 mm accelerates the distillation process at a temperature of 150 °C almost twice. During the experiments, when the temperature rises above 190 °C, there is a deterioration in the consumer properties of the essential oil, *i.e.* a change in smell occurs. In this regard, a temperature limit is found in which there is no decrease in the quality characteristics of the product. To solve these problems, an additional series of experiments was set. As the studied parameters, the distillation duration and the temperature of the working steam were chosen. Experimental verification confirmed the effectiveness of this equation. As it can be seen, the optimum temperature for distillation of the essential oil is (190 ± 3) °C. It is increase that not only leads to a decrease in prod-

uct yield but also worsens its quality characteristics.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

References

- [1] Semerdjieva, B.I., Shiwakoti, S., Cantrell, L.C., Zheljzkov, D.V., Astatkie, T., Schlegel, V. and Radoukova, T. (2019) Hydrodistillation Extraction Kinetics Regression Models for Essential Oil Yield and Composition in *Juniperus virginiana*, *J. excelsa*, and *J. sabina*. *Molecules (Basel, Switzerland)*, **24**, 986. <https://doi.org/10.3390/molecules24050986>
- [2] Sela, F., Karapandzova, M., Stefkov, G., Cvetkovikj, I. and Kulevanova, S. (2015) Chemical Composition and Antimicrobial Activity of Essential Oils of *Juniperus excelsa* Bieb. (Cupressaceae) Grown in R. Macedonia. *Pharmacognosy Research*, **7**, 74-80. <https://doi.org/10.4103/0974-8490.147212>
- [3] Severoglu, Z., Ozyigit, I.I., Dogan, I., Kurmanbekova, G., Demir, G., Yalcin, I.E. and Kari, G.K. (2015) The Usability of *Juniperus virginiana* L. as a Biomonitor of Heavy Metal Pollution in Bishkek City, Kyrgyzstan, *Biotechnology & Biotechnological Equipment*, **29**, 1104-1112. <https://doi.org/10.1080/13102818.2015.1072478>
- [4] Nikolic, B., Tesevic, V., Djordjevic, I., Jadranin, M., Bojovic, S. and Marin, P.D. (2009) *n*-Alkanes in Needle Waxes of *Picea omorika* VAR. *vukomanii*. *Chemistry of Natural Compounds*, **45**, Article No. 697. <https://doi.org/10.1007/s10600-009-9430-y>
- [5] Radulescu, V., Saviuc, C., Chifiriuc, M. and Oprea, E. (2011) Chemical Composition and Antimicrobial Activity of Essential Oil from Shoots Spruce (*Picea abies* L). *Revista de Chimie Bucharest*, **62**, 69-74.
- [6] Haman, N., Morozova, K., Tonon, G., Scampicchio, M. and Ferrentino, G. (2019) Antimicrobial Effect of *Picea abies* Extracts on *E. coli* Growth. *Molecules (Basel, Switzerland)*, **24**, 4053. <https://doi.org/10.3390/molecules24224053>
- [7] Modnicki, D. and Łabędzka, J. (2009) Estimation of the Total Phenolic Compounds in Juniper Sprouts (*Juniperus communis*, Cupressaceae) from Different Places at the Kujawsko-Pomorskie Province. *Herba Polonica*, **55**, 127-131.
- [8] Fierascu, I., Ungureanu, C., Avramescu, S.M., Cimpeanu, C., Georgescu, M.I., Fierascu, R.C., Ortan, A., Sutan, A.N., Anuta, V., Zangfirescu, A., Dinu-Pirvu, C.E. and Velescu, B.S. (2018) Genoprotective, Antioxidant, Antifungal and Anti-Inflammatory Evaluation of Hydroalcoholic Extract of Wild-Growing *Juniperus communis* L. (Cupressaceae) Native to Romanian Southern Sub-Carpathian Hills. *BMC Complementary and Alternative Medicine*, **18**, Article No. 3. <https://doi.org/10.1186/s12906-017-2066-8>
- [9] Banerjee, S., Mukherjee, A. and Chatterjee, T.K. (2012) Evaluation of Analgesic Activities of Methanolic Extract of Medicinal Plant *Juniperus communis* Linn. *International Journal of Pharmacy and Pharmaceutical Sciences*, **4**, 547-550.
- [10] Almalki, D.A., Alghamdi, S.A. and Al-Attar, A.M. (2019) Comparative Study on the Influence of Some Medicinal Plants on Diabetes Induced by Streptozotocin in Male Rats. *BioMed Research International*, **2019**, Article ID: 3596287. <https://doi.org/10.1155/2019/3596287>
- [11] Burits, M., Asres, K. and Bucar, F. (2001) The Antioxidant Activity of the Essential

- Oils of *Artemisia afra*, *Artemisia abyssinica* and *Juniperus procera*. *Phytotherapy Research*, **15**, 103-108. <https://doi.org/10.1002/ptr.691>
- [12] Miceli, N., Trovato, A., Dugo, P., Cacciola, F., Donato, P., Marino, A., Bellinghieri, V., Barbera, T.M.L., Güvenç, A. and Taviano, M.F. (2009) Comparative Analysis of Flavonoid Profile, Antioxidant and Antimicrobial Activity of the Berries of *Juniperus communis* L. var. *communis* and *Juniperus communis* L. var. *saxatilis* Pall. From Turkey. *Journal of Agricultural and Food Chemistry*, **57**, 6570-6577. <https://doi.org/10.1021/jf9012295>
- [13] Akdogan, M., Koyu, A., Ciris, M. and Yildiz, K. (2012) Anti-Hypercholesterolemic Activity of *Juniperus communis* Lynn Oil in Rats: A Biochemical and Histopathological Investigation. *Biomedical Research*, **23**, 321-328.
- [14] Tavares, L.R. (2012) Evaluation of Neuroprotective Potential of Polyphenols Derived from Portuguese Native Plants: *Juniperus* sp. and *Rubus* sp. PhD Thesis, Universidade Nova de Lisboa, Lisbon, 256 p.
- [15] Öztürk, M., Tümen, I., Uğur, A., Aydoğmuş, O.F. and Topçu, G. (2011) Evaluation of Fruit Extracts of Six Turkish *Juniperus* Species for Their Antioxidant, Anticholinesterase and Antimicrobial Activities. *Journal of the Science of Food and Agriculture*, **91**, 867-876. <https://doi.org/10.1002/jsfa.4258>
- [16] Karlsen, J. and Svendsen, A.B. (2002) The Influence of the Isolation Method on the Composition of the Essential Oil of Leaves and Twigs of *Juniperus communis* L. var. *saxatilis* Pall Growing in Norway. *Scientia Pharmaceutica*, **70**, 87-92. <https://doi.org/10.3797/scipharm.aut-02-10>
- [17] Naji, G., Mellouk, H., Rezzouget, S.A. and Allaf, K. (2008) Extraction of Essential Oils of Juniper Berries by Instantaneous Controlled Pressure-Drop: Improvement of DIC Process and Comparison with the Steam Distillation, *Journal of Essential Oil Bearing Plants*, **11**, 356-364. <https://doi.org/10.1080/0972060X.2008.10643641>
- [18] Benchaar, C., Calsamiglia, S., Chaves, A.V., Fraser, G.R., Colombatto, D., McAllister, T.A. and Beauchemin, K.A. (2008) A Review of Plant-Derived Essential Oils in Ruminant Nutrition and Production. *Animal Feed Science and Technology*, **145**, 209-228. <https://doi.org/10.1016/j.anifeedsci.2007.04.014>
- [19] Radoukova, T., Zheljzakov, V.D., Semerdjieva, I., Dincheva, I., Stoyanova, A., Kačaniová, M., Marković, T., Radanović, D., Astatkie, T. and Salamon, I. (2018) Differences in Essential Oil Yield, Composition, and Bioactivity of Three Juniper Species from Eastern Europe. *Industrial Crops and Products*, **124**, 643-652. <https://doi.org/10.1016/j.indcrop.2018.08.012>
- [20] Dudchenko, L.G., Koz'yakov, A.S. and Krivenko, V.V. (1989) Spicy-Aromatic and Spicy-Flavoring Plants: Handbook. Naukova Dumka, Kyiv, 304 p.
- [21] Serbin, A.G., Seraya, L.M., Tkachenko, N.M. and Slobodnyk, T.A. (2003) Medical Botany. Textbook for University Students. Publishing house of National Pharmaceutical University, Kharkov, 364 p.
- [22] Gubanov I.I., Krylova I.T. and Tikhonova V.L. (1976) Wild useful plants of the USSR. In: Rabotnov, T.A., Ed., *Reference Guides-Identifiers of a Geographer and a Traveler*, Mysl, Moscow, 107-108.
- [23] Mashkovsky, M.D. (1972) Medicines. 2nd Edition, Medicine, Moscow, 486 p.