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Determination of Methanol and Ethanol Levels in Herbal Distillates (Mint, Burdock, Spring Orange, Chicory, Thyme, Chives, and Horseradish) at Supply Level of Ahvaz Using Gas Chromatography

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ABSTRACT

Introduction: The risk of poisoning and death with oral consumption of ethanol and methanol in food is of particular importance. In this study, seven types of commonly used herbal distillates were purchased through random sampling from stores at the supply level of Khuzestan for testing.

Methods: In this research paper, the levels of ethanol and methanol in herbal distillates of mint, burdock, spring orange, chicory, thyme, chives, and horseradish were determined using gas chromatography. Each sample was measured three times, and the numerical average was reported.

Results: The average ethanol levels for the herbal distillates of mint, burdock, spring orange, chicory, thyme, chives, and horseradish were 7.36, 159.17, 12.66, 4.24, 4.53, 38.66, and 8.82 ppm, respectively. Additionally, the average methanol levels for the herbal distillates of mint, burdock, spring orange, chicory, thyme, chives, and horseradish were 1, 2, 3, 4, 5, 6, and 7, respectively.

Conclusion: The studied herbal distillates contain ethanol and methanol. Excessive and excessive use of these traditional products can lead to poisoning, especially in children and individuals with special conditions such as pregnant women and chronic patients.

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Introduction

From ancient times, humans often used plants for the treatment of diseases; in fact, they turned to nature to be relieved from pain and various ailments (Picking, 2024). People who use natural and non-chemical substances for the treatment of diseases usually use different parts of plants, including roots, stems, leaves, and even their bark in their work (Picking, 2024). Herbal medicine was commonly used as a treatment method, and with the development of herbal medicine technology and the widespread use of these products, it is becoming mainstream in the field of medicine. This is because improvements in analytical and quality control processes,

along with advances in clinical research, have demonstrated the value of herbal medicines in the treatment and prevention of diseases (Khadka et al., 2021). Medicinal plants have been used for medicinal purposes long before the recording of history. Various forms of medicinal plants are used as medicinal products. Herbal ointments, herbal tablets and capsules, syrups, supplements. Another form in which medicinal plants can be used as medicines for treating diseases is the preparation of herbal distillates (Bhowmik et al., 2009).

Herbal distillates are traditional herbal products. Herbal distillates contain medicinal properties because they contain chemical compounds, active ingredients, and antioxidants (Hosseini et al., 2023). Herbal distillates are one of the simple and practical methods for preparing traditional medicines from which has gained popularity today. Herbal distillates are actually obtained from the distillation of the essence of different parts of plants (Hosseini et al., 2023). Medicinal distillates are actually the fragrance, perfume or essence of plants that are added to water. They are fragrant medicines whose scents can stimulate the olfactory nerve receptors and affect the production of hormones, body temperature, metabolism, stress and sex hormone levels, the body's defense system, our emotional and physical thoughts and behaviors. These fragrant substances, which are more abundant in herbal extracts or oils, stimulate the brain and release neurotransmitters, leading to specific mental states in humans and creating a sense of health, joy, pleasure, and happiness (Ghorbani et al., 2023). Unlike chemical drugs, herbal medicines and distillates are relatively safe for human use and are environmentally friendly. Herbal distillates are also economically affordable and easily accessible (Qadi Pasha et al., 2022). Due to the side effects of chemical drugs, people nowadays tend to prefer herbal medicines and distillates (Qadi Pasha et al., 2022). The medicinal properties of herbal distillates are numerous, including benefits for the skin, obesity or weight loss, nerves, stomach, blood thinning or blood pressure reduction, stomach strengthening, heart and liver strengthening, deworming and parasite removal, liver and stomach cleansing, sleep induction, stress relief, and more (Qadi Pasha et al., 2022). During the production process of herbal distillates, for the extraction and procurement of stronger herbal distillates, first, second, or third-class alcoholic solvents such as ethanol, methanol, etc., are sometimes used, and occasionally in the alcohol removal or other physiological processes of the plant, alcohol is produced and enters the herbal distillate product (Hosseini et al., 2023). Due to the high consumption of herbal distillates; despite their important medicinal effects, these herbal distillates also have their side effects and are not all suitable (Qadi Pasha et al., 2022). In the production process of herbal distillates, alcoholic solvents (ethanol, methanol, etc.) are used, and it is possible that their residues remain in the herbal distillates. The health of these widely consumed natural products is of great importance, and therefore, testing and examining these products for the presence of alcohols such as methanol and ethanol, which have severe side effects, is of particular importance.

Therefore, the aim of this study was to investigate the levels of methanol and ethanol in herbal distillates such as mint, burdock, spring orange, chicory, thyme, chives, and horseradish using gas chromatography.

Materials and Methods

Sample Collection

In this study, common herbal distillates used in the city of Ahvaz (Iran) are including Mint, burdock, spring orange, chicory, thyme, chives and horseradish were collected and sampled.

Sample Size Calculation Method

Three samples of each type of herbal distillates were selected at random from the stores of the supply levels of Ahvaz city. The herbal distillates that were tested included burdock, spring orange, chicory, thyme, chives and horseradish.

Conditions and Method of Chromatography

The concentrations of ethanol and methanol were determined using a model Agilent 7890B GC instrument with an FID detector. The column used was RTX-5MS, 30 m in length, 0.25 mm internal diameter, and 0.25 µm thickness of stationary phase. The initial temperature of the device was 40 ºC, which was maintained for 3 minutes, and then it reached the temperature of 160 ºC with a ramp of 20 ºC per minute and was maintained at this temperature for 1 minute. Injections were made in splits with a ratio of 1:25. The injection volume was 1 microliter. The temperature of the injector was 200 ºC and the temperature of the detector was 250 ºC. Due to the transparency of the samples, the 1 microliter sample volume was diluted in splits with a ratio of 1:25. This means that the apparatus divided the sample into 25 parts and 1 part of the sample was analyzed and 24 parts were taken out of the apparatus. Nitrogen was also used as the carrier gas for the make-up.

Results

In this study, 21 samples of herbal distillates from 7 types of herbal distillates were selected, and the amount of methanol and ethanol present in them was investigated. The results of our study showed that all herbal distillates contained ethanol and methanol. The results of ethanol and methanol levels are given in Table 1.

Table 1: Mean levels of ethanol and methanol in herbal distillates of mint, burdock, spring orange, chicory, thyme, chives and horseradish.

Herbal distillates	Average ethanol (ppm)	Average methanol (ppm)
Mint	7.36	16.26
Burdock	159.17	65.37
Spring orange	12.66	12.61
Chicory	4.24	7.97
Thyme	4.53	36.55
Chives	38.66	11.33
horseradish	8.82	35.24

Discussion

Herbal distillates are one of the traditional medicinal products and are widely consumed among people. Given the great importance of herbal distillates and the various medicinal uses of different types of traditional and industrial distillates, their chemical and microbial health and safety are very important. Studies show that blurred vision leading to blindness following the unintentional consumption of methanol along with the use of some herbal distillates has caused concern among health authorities and sensitivity of health officials. The results obtained from the study showed that, regardless of the manufacturing plant, all the herbal distillates in our study contained varying amounts of methanol and ethanol (Nekoukar et al., 2021). The presence of alcohols in herbal distillates has been reported in a few studies. The results of a study by Salahi et al. (2009) showed that the highest concentration of methanol was related to the industrially produced peppermint distillate, which was 415.04 ppm, and the lowest was related to a sample of handmade rose water distillate at 60.26 ppm. Statistically, there was no significant difference between the concentration of methanol in handmade herbal distillates and industrially produced herbal distillates (Solhi et al., 2009). The results of another study showed that the bulk herbal distillates in the Tehran (Iran) market were free of ethanol and methanol. The level of methanol in peppermint distillate was 334.62±153 ppm, in rose water distillate 321.4 ppm, and in tarragon distillate 311.39 ppm, and pennyroyal distillate was free of methanol (Ghadipasha et al., 2022). The results of a study showed that the highest level of methanol was in yarrow water containing 1477.7±23.8 ppm and burdock distillate containing 79.4±3 ppm methanol (Karimi et al., 2008). The results of a study by Mousavi et al. (2011) showed that the highest concentration of methanol was in yarrow water containing 1208±74.202 ppm milligrams per liter, and cinnamon distillate also contained 18.93±1.04 mg/l methanol (Mousavi et al., 2011). The results of a study by Rafiee Zadeh et al. showed that milk thistle distillate contained 310 mg/l and rose water contained 65 mg/L (Rafizadeh et al., 2015). According to American standards, the amount of methanol in fresh and canned fruit juices such as grapefruit and orange is 12-64 mg/L (Committee on toxicology, 2008). Stressors such as increased ozone concentration, hypoxia due to snowfall, leaf drying, and aging of plant tissues cause methanol production in the plant (Jacob et al., 2004).

There are many diverse factors that can affect the amount of methanol in an herbal distillate, the most important of which are the origin of the plant tissue used in distillation, genetic and racial differences of plants, climatic and weather conditions, the age of the plant used, the distillation method, and so on (Nonomura and Benson, 1992). Usually in the distillation of plants, to increase the work efficiency and improve the quality of the product, the plant or flower is picked after about 48 hours of water stress and used. This action, as a kind of stress, increases the concentration of methanol in the plant or flower and consequently in the resulting distillate, which can justify the presence of methanol in products.

The simplest single-carbon alcohol is methanol (Nonomura and Benson, 1992). Methanol produced in plants is stored in the water inside the tissues and also in some plant tissues, and some of it is initially converted to formaldehyde and then to formic acid and finally to CO2 inside the plants (Hemming et al., 1995). Methanol is oxidized in the plant in the form of formaldehyde and CO2 and synthesized in the form of carbohydrates and amino acids, including serine and methionine, in various plant tissues as 3C (Nonomura and Benson, 1992). Studies show that spraying methanol on three-carbon plants is effective in increasing yield, reducing water needs, uniform maturation, and reducing the effect of drought stress on plants (Sawan et al., 2001). On the other hand, methanol produced inside plants, in addition to its direct effect on their growth, also has significant effects on plant growth through stimulating the activity of methylotrophic bacteria (Holland et al., 2002; Abanda-Nkpwatt et al., 2006). Published and scattered reports from some physicians about the presence of symptoms of methanol poisoning, including blindness, in chronic

consumers of herbal distillates indicate the importance of measuring methanol in such products (Karimi et al., 2007). Recently, reports of cases of blurred vision leading to blindness following the unintentional consumption of methanol along with the use of some herbal distillates have caused serious concern and sensitivity among health officials in the country. Unlike alcoholic beverages, part of the methanol in herbal distillates is derived from the plant's metabolic processes during growth and even after harvesting the plant until the distillate is prepared (Rafizadeh et al., 2010; Rafizadeh et al., 2011; Rafizadeh et al., 2013). The presence of ethanol in herbal distillates is due to the addition of this alcohol during distillation as a solvent for better extraction of the herbal distillate. Methanol is a highly toxic aliphatic alcoholic compound that is used in industry as a solvent and in the production of formaldehyde and methylated compounds. Methanol exists as an impurity in many processed plant-based foods, and its maximum permissible concentration in alcoholic beverages is 200 parts per million, and this number should be lower in non-alcoholic products (Aine and Davan, 2007). Ingested methanol is completely absorbed and reaches its maximum serum concentration within 30 to 60 minutes after ingestion, and methanol poisoning may occur (Ford et al., 2000). Therefore, continuous and excessive consumption of herbal products (herbal distillates) and prolonged use of them can lead to poisoning. Although technical factors can influence the production of products with varying amounts of alcohols, especially methanol, in different factories, physiological reactions and the addition of alcoholic solvents are also factors influencing the presence of methanol and ethanol in herbal distillates. Since herbal distillates such as peppermint, burdock, spring orange, chicory, thyme, parsley, and milk thistle are widely used among people, it is necessary for food and beverage control laboratories to consider periodic testing of these products from the factory gate to provide a safe and secure product to the consumer.

Conclusions

Some types of distillates may be used in large amounts for a short period to treat certain diseases, and there is a risk of methanol poisoning in such cases. Therefore, raising public awareness and determining the permissible level of methanol in herbal distillates is an important step in preventing such cases.

Declarations

Conflict of interest

No conflict of interest among the authors.

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Consent for publications

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Authors' contributions

MP contributed in conceptualization, methodology, software, validation, formal analysis, investigation, resources, data curation, original draft preparation, review and editing, visualization, supervision, and project administration. MP contributed in conceptualization, methodology, investigation, resources, review, and editing. DA contributed in conceptualization, original draft preparation, review, and editing.

Ethical considerations

Ethical issues (including plagiarism, misconduct, data fabrication, falsification, double publication or submission, redundancy) have been completely observed by the authors.

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