Holistic utilization of oregano: optimization of essential oil distillation and valorisation of distilled oregano waste

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Origanum species are worldwide distributed herbs that belong to *Lamiaceae* family and are primarily cultivated in Eurasia and Mediterranean zone (Lukas *et al* 2013). Their uses are extended as spices in food, pharmaceutical, and cosmetics industry. *Origanum vulgare* ssp. *Hirtum* is one of the most important species and it is also called "Greek oregano" as it is native to many Greek areas. According to Goliaris and Skroubis (1992), "Greek oregano" is characterized by a high concentration of exceptional essential oil. Oregano oil has long been used in traditional medicine for its antimicrobial, anti-inflammatory, antifungal, and antioxidant properties. It is rich in the monoterpenic phenol carvacrol and its isomeric analog, thymol.

Considering that oil quality is related to its chemical composition, the choice of a suitable extraction method is extremely important. Hydrodistillation is one of the most used methods for extraction of essential oils from plants and it is carried out using a Clevenger apparatus. Although conventional methods are most commonly used to extract essential oil from plants, they have some drawbacks, such as the difficultly to control the heat transfer throughout the process and the long extraction time. Moreover, many natural products are thermally unstable and can be damaged during thermal extraction. In this regard, the need for novel extraction methods has become more intense. The extraction of essential oils using an ultrasound-assisted process has been recommended by several authors as one of the most efficient extraction system presenting many benefits, such as high extraction yield, low energy consumption and processing time, high reproducibility (Wen *et al* 2020). Ultrasound effect is based on cavitation phenomenon (Toma *et al* 2001). The implosion of cavitation bubbles on a product surface results in micro-jetting that leads to several effects such as surface peeling, erosion, and particle breakdown, whereas implosion of cavitation bubbles in a liquid generates macro-turbulences and a micro mixing (Alexandru *et al* 2013). Many researchers have used an ultrasound treatment before hydrodistillation to extract essential oils from herbs such as *Cinnamomum cassia*, *Aloysia citriodora Palau*, *Prangos ferulacea Lindl.*, *Satureja macrosiphonia Bornm*, and *Daucus carota*. In addition, the ultrasound-assisted hydrodistillation has been studied for herbs such as *Cymbopogon winterianus*, *Iberis amara*, and *Allium sativum*. However, no reports have been found on the use of ultrasounds in the extraction of oregano oil.

In general, the essential oils industry generates large amounts of wastes due to the low content of oil in the fresh plants. Most of the distilleries simply discard the waste biomass, which could lead to ecological problems in the nearby locations. Various methods for this waste utilization have been successfully developed during the past years. Apart from the common methods of disposal (composting, use for animal feed or simply discarding), there are several other promising and widely investigated strategies, such as recovery of valuable biologically active substances and their application to food industry.

The objective of this work is the holistic utilization of oregano through a) the optimization of essential oil hydrodistillation using ultrasounds pretreatment or ultrasound-assisted hydrodistillation and b) the valorization of distilled oregano waste for the recovery of phenolic compounds.

Origanum vulgare ssp. *Hirtum* essential oil was obtained using three different methods of distillation, conventional hydrodistillation (CH), hydrodistillation with ultrasound application (UH), and hydrodistillation with ultrasound pretreatment (UPH). For each method, various parameters, such as amplitude level, pulse duration/pulse interval ratio, and treatment time, were examined for their effect on oil yield. The optimum distillation method and operating conditions were determined. In all treatments, the oil was collected at various time intervals and two mathematical approaches were selected for the kinetic modelling, namely, the first order kinetic model and the Peleg's model. Different parameters were analyzed for their effects on kinetic models constants.

The distilled oregano waste obtained at the optimum conditions of each method was oven-dried and used for phenolic compounds extraction using different solvents (50 and 70% ethanol, 50 and 70% methanol). Conventional maceration and ultrasound-assisted extraction were the two investigated methods. Total phenolic content was estimated spectrophotometrically according to the Folin-Ciocalteu colorimetric method.

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