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One stage olive mill waste streams valorisation via hydrothermal carbonisation

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ABSTRACT

An olive waste stream mixture, coming from a three phase-continuous centrifugation olive oil mill industry, with a typical wet basis mass composition of olive pulp 39 wt%, kernels 5 wt% and olive mill waste water 56 wt%, was subjected to hydrothermal carbonisation (HTC) at 180, 220 and 250 °C for a 3-hour residence time in a 2-litre stainless steel electrically heated batch reactor. The raw feedstock and corresponding hydrochars were characterised in terms of proximate and ultimate analyses, higher heating values and energy properties. Results showed an increase in carbonisation of samples with increasing HTC severity and an energy densification ratio up to 142% (at 250 °C). Hydrochar obtained at 250 °C was successfully pelletised using a lab scale pelletiser without binders or expensive drying procedures. Energy characterisation (HHV, TGA), ATR-FTIR analysis, fouling index evaluation and pelletisation results suggested that olive mill waste hydrochars could be used as energy dense and mechanical stable biofuels. Characterisation of HTC residues in terms of mineral content via induced coupled plasma optical emission spectroscopy (ICP-OES) as well as Total and Dissolved Organic Carbon enabled to evaluate their potential use as soil improvers. Nutrients and polyphenolic compounds in HTC liquid fractions were evaluated for the estimation of their potential use as liquid fertilisers. Results showed that HTC could represent a viable route for the valorisation of olive mill industry waste streams.

1. Introduction

Efficient valorisation of agricultural and agro-industrial waste is a key factor in developing new strategies for the circular economy. The direct use of such residues as soil improvers and/or biocombustibles is often hampered by their phytotoxicity, high moisture content, high biodegradability and low energy density. Furthermore, olive oil production takes place for a short period of the year (typically between October and December). The disposal of olive mill waste is thus also complicated due to the seasonality of their production. High amounts of organic waste accumulated in a short period of time impose high removal costs on companies and pose an environmental risk arising from the still widespread use of improper disposal practices. For those reasons the development of innovative approaches to the conversion of wet waste biomass into valuable materials, such as stable solid bio-fuels and/or soil improvers, represents a priority in the agro-industry sector and in particular in biological and conservation agriculture (Li et al., 2018). In particular, the olive mill industry could benefit from the development of a technology capable of eliminating the by-products and converting them into valuable resources. Worldwide virgin olive oil production in 2017 was estimated at around 2.84 Mt (World Olive Oil Production, 2017). Virgin olive oil is largely produced in the Mediterranean area, which accounts for more than 87 wt% of the world production (FAOSTAT, 2014). In 2014 the largest olive oil producers' countries (Spain, Italy, Greece, Tunisia, Morocco, Turkey and Algeria) accounted for 58.0, 9.8, 7.0, 5.7, 4.6, 2.5 and 1.7 wt% of the total world production, respectively (FAOSTAT, 2014). Considering that, on average, virgin oil production amounts between 15 and 20 wt% of fresh olives milled, olive mill waste approximately ranges between 80 and 85% of the total feedstock weight, or even more when additional water is used during milling (FAOSTAT, 2014). Based on the above, it can be estimated that in the Mediterranean region more than 15 Mt of olive waste are produced between October and December each year. The separation and trading of olive kernels (approximately 5 wt%

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of the total wet residue) and oil extraction from olive pulp and kernels to produce olive pomace oil (an additional 5–6 wt%) have been almost abandoned due to their high process and transportation costs. The direct use of olive mill waste water (OMWW) as soil improver is limited due to its high concentration of polyphenolic species, which could prove toxic to the ecosystems. Agronomic use of OMWW (Kavvadias et al., 2010) and olive mill solid waste on agricultural land may affect soil properties and subsequently enhance the risk for groundwater contamination (Chowdhury et al., 2015). Thus numerous studies have recently focused on the recovery and valorisation of those residues. Olive solid waste treatments are based on agronomic, chemical, biochemical and thermochemical processes (Christoforou and Fokaides, 2016). One of the most followed approaches for olive mill waste effluent treatments is anaerobic digestion (AD). Despite recent advances in AD technology (Gunay and Karadag, 2015), the anaerobic treatment of olive mill waste residues represents a challenge due to the high content of polyphenols compounds which inhibit biogas formation (Chen et al., 2008). To overcome the inhibition process, mixing with different types of waste is necessary (Surra et al., 2017; Pellera and Gidarakos, 2017; Maragkaki et al., 2018). Dry thermochemical treatments for energy recovery from food waste, municipal solid waste and olive mill solid residues have been also extensively investigated (Gopu et al., 2018, Lombardi et al., 2015; Volpe et al., 2014; Messineo et al., 2012). Torrefaction of olive pulp residues, olive pruning and kernels was used as a pre-treatment for the production of a more brittle and stable bio-fuel (Benavente and Fullana, 2015; Volpe et al., 2016) and slow pyrolysis for the production of high energy density material (Volpe et al., 2015). More recently, high performance membrane extraction technologies have been developed (Gebreyohannes et al., 2016; Bazzarelli et al., 2016). Membrane technologies have been used to overcome the high risk associated with OMWW disposal and valorise the olive mill by-products by converting them into organic fertilisers (Galliou et al., 2018) or valuable chemicals (i.e. polyphenols compounds). High investment and operating costs, operational challenges due to membrane fouling and difficulties in efficient polyphenols separation and recovery still represent a severe limitation in the development of OMWW membrane separation technology (Pulido, 2016).

Among the different biomass conversion technologies, wet thermochemical processes as HTC have recently attracted great interest in the scientific community (Lucian et al., 2018). That is due to the possibility of direct conversion of high moisture biomass into valuable bio-fuels, which would avoid expensive drying pretreatments. During HTC, wet biomass is heated up in subcritical water at temperatures up to 280 °C and pressures equal to or higher than those of saturated steam. The dramatic change in water properties (i.e. dielectric constant and polarity) catalyses the hydrolysis of ligno-cellulosic materials (Kruse et al., 2013).

A recent study evaluating energy efficiency and cost analysis of conversion of waste biomasses via HTC, followed by hydrochar pelletisation, showed that HTC could represent an economic and sustainable way for the production of solid densified bio-fuels (Lucian and Fiori, 2017). The mechanical strength, low moisture uptake and improved combustion properties indicated that hydrochar pellets are more suitable solid biofuels than raw biomass pellets (Liu et al., 2014). Pilot plants installations proved that HTC technology could be economically operated and easily scaled and could serve as a local bio-refinery (Hitzl et al., 2015). Several studies addressing HTC of olive mill waste have been recently published. HTC of OMWW led to low hydrochar yields due to a scarce content of carbohydrates in the feedstock (Poerschmann et al., 2013a). However, it also revealed the possibility of a more easy extraction of valuable phenolic species from the HTC liquid residue when compared to the starting material (Poerschmann et al., 2013b). HTC of olive kernels (Donar et al., 2016) and olive pomace (Volpe and Fiori, 2017; Missaoui et al., 2017; Benavente et al., 2015) was found to produce high energy density hydrochars with potential use as bio-fuel feedstock. Recently, a life cycle analysis of olive mill effluents revealed that HTC could be a more environmentally favourable technology when compared with anaerobic digestion and incineration, if liquid HTC residues are also appropriately valorised (Benavente et al., 2017). Differently to what already reported, this study reports the results of HTC treatment of three-phase olive mill waste effluents, including OMWW. Although many technologies have been investigated and partially developed for the treatment of the different olive oil mill industry effluents, none of them has up to now offered an easy, cost-efficient and viable way to treat all waste streams (liquids and solids) at the same time, without high investment costs and/or high costs associated to residue transportation to the process facilities. The "one stage" HTC process reported in this study could prove to be a "greener" alternative, an easy scalable technology that would fulfil the mill needs (i.e. quantity and nature of effluents) and enable the simultaneous processing of all waste effluents, thus producing a high energy biofuel and a soil improver at the same time.

To the best of our knowledge, this is the first example where the entire waste stream of an olive mill factory is used simultaneously in a HTC process for the production of a solid biofuel and a soil improver.

2. Material and methods

2.1. Materials and sample preparation

Olive oil mill waste (OMW) including olive pulp (OP), olive kernels (OK) and olive mill waste water (OMWW) were provided by Agririva company, a typical three-phase decanter olive mill factory located in Riva del Garda (TN), Italy. The waste materials (both solid and liquid) were stored in a freezer at −20 °C to preserve their original characteristics, and defrosted just before each experiment. OP and OK were dried in a convection drying oven at 105 °C before characterisation analysis. Analysis carried out on raw OMWW was performed before freezing. Moisture content of OP and OK were measured following EN ISO 18134-3:2015 standard method and resulted equal to 65.7 ± 0.3 wt% and 20.0 ± 0.2 wt%, respectively. All samples were defrosted overnight and used without further treatment prior to HTC tests.

2.2. HTC system set-up and procedures

The HTC experiments were performed in an electrically heated 2000-mL stainless steel (AISI 316) batch reactor, under controlled temperature regimes, specifically designed and manufactured at the University of Trento (Merzari et al., 2018). For each experiment, 528 ± 0.1 g of wet OP sample (about 180 g on a d.b.) were mixed with 72 ± 0.1 g of wet OK (about 57 g on a d.b.) and placed in the reactor. Subsequently, 750 ± 0.1 g of OMWW (containing about 7 wt% of SS) were weighed and added to the reactor to obtain the olive waste mixture (OWM) respecting the exact proportion of waste produced by a typical three-phase decanter olive mill factory during virgin olive oil extraction. Once sealed, the reactor was flushed with pure nitrogen (N_2, AirliquideAlphagaz™1) and the system heated up to the set temperature (temperature gradient of about 10 °C min^{-1}). Reaction peak temperature was kept for a fixed 3-hour residence time for all the experiments. After the desired reaction time had elapsed, the reaction was quenched by cooling the reactor through a cold (−30 °C) stainless steel plate placed at its bottom. Meanwhile, compressed air was blown to the reactor walls to speed up the cooling process. When the reactor's temper-