Perspective

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The citrus economy in Sicily in the early bioeconomy era: a case study for bioeconomy practitioners

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Abstract: Sicily hosts a significant and ancient citrus agroindustry, which includes more than 42 000 farming companies and around 20 citrus fruit-processing companies. This study critically analyzes the citrus economy in Sicily in the early bioeconomy era with the aim to identify outcomes of general relevance for bioeconomy researchers and bioeconomy company managers working towards the full valorization of *Citrus* crops. These practitioners include bioeconomy company managers who need additional education in which economic and management knowledge is merged with an understanding of science and technology. © 2024 The Authors. *Biofuels, Bioproducts and Biorefining* published by Society of Industrial Chemistry and John Wiley & Sons Ltd.

Key words: citrus; bioeconomy; Sicily; orange; pectin; lemon

Introduction

With a planting area exceeding 88 000 ha and 42 500 farms, Sicily hosts the largest citrus fruit (genus *Citrus*; family Rutaceae) cultivations in Italy.[1] About 58 000 ha of land are cultivated with orange (*Citrus sinensis*), 21 000 ha with lemon (*C. limon*), and 5000 ha with mandarin (*C. reticulata*). The annual overall production of citrus fruits, which also includes clementines (*C. clementina*) and grapefruit (*C. paradisi*), amounts to about 1.47 million tonnes (1 million t oranges, 0.4 million t lemons, 60 000 t mandarins, and 50 000 t clementines, with minor amounts of grapefruits and tangerines).[2] In 2000, for comparison, the overall cultivated land was 106 644 ha.[3]

Revenues exceed €532 million, with a 31 000 workforce (including employees in commercialization),[1] but production is highly fragmented: 3 ha is the average area cultivated by a single farm.[1] About one third of orange fruits are squeezed by the citrus industry in Sicily to produce fresh or concentrated orange juice (and orange essential oil) generating 340.000 t of waste orange peel (WOP), which is mostly discarded as waste at substantial economic cost.[4]

Apart from a water content of 85%, the WOP dry part chiefly consists of pectin (42.5 g/100 g), sugars (16.9 g/100 g), cellulose (9.21 g/100 g), and hemicellulose (10.5 g/100 g).[5] Other valued bioproducts include essential oils (EOs), phenolic compounds, vitamins and organic acids, including citric acid, which for decades was produced from lemon juice.[6]

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Many studies between 2002 and 2022 were devoted to WOP valorization.[7,8] At the end of the 2010s, Thompson and co-workers first wrote about the 'biorefinery of waste orange peel'.[9] Recently, Guo and co-workers reviewed numerous studies in which fermentation bioconversion technologies were employed to convert waste orange peel to valued bioproducts such as lactic acid, succinic acid, xanthan gum, and biofuels (methane and ethanol).[10] Starting from orange peel hydrolysate (shredded orange peel is mixed with water in 1:2 weight ratio, added with 0.5–2.5% sulfuric acid and briefly hydrolyzed at 121°C), the direct production of valued xanthan gum (sold at USD 5000/t) over strains of *Xanthomonas campestris* is particularly promising.[11]

A Boolean search carried out in early October 2023 on a research database with the queries 'orange peel' and 'valorization' returned 5781 documents, of which 3745 were research papers.[12] Zuin and co-workers in Brazil, by far the world's largest orange crop-growing country, have even designed a laboratory practice for chemistry and chemical engineering students to illustrate the main concepts of green chemistry through a greener bioproduct extraction method applied to citrus waste (a redesigned distillation system in which the steam generator is separated from the apparatus containing the biowaste).[13]

The main current use of WOP as a biochemical raw material is in conventional pectin extraction with mineral acid. The hydrolytic extraction process followed by vacuum evaporation and alcohol precipitation using large quantities of expensive and flammable alcohol, however, has both high capital and operational costs.[14]

Critically analyzing the citrus economy in Sicily in the early bioeconomy era, this study aims to identify outcomes of general value that may assist the work of bioeconomy practitioners engaged in the full valorization of *Citrus* crops in countries where the genus is widely cultivated. These practitioners include bioeconomy company managers who need additional education in which economic and management knowledge is merged with an understanding of science and technology.[15]

Following a brief overview of the story of citrus cultivation in Sicily (and in Italy), we discuss selected green chemistry and bioeconomy aspects presenting arguments from a practical (economic and industrial) perspective.

The citrus economy in Sicily

The citrus tree originates from the subtropical and tropical regions of Asia and the Malay Archipelago.[16] It was well known to the Ancient Greeks and the Romans (in his *Naturalis Historia* of 77 AD Pliny named it *malus medica*, *malus assyria*, and *citrus*) and it was cultivated by them in southern Italy at least for ornamental reasons (as proven by the seeds found amid the Pompei not far from a tile mosaic reproducing the orange tree).[17]

From a vaulted ceiling mosaic in Rome, designed in about 330 AD for the grave of Constantia, daughter of Constantine the Great, reproducing lemon, citron and orange fruits attached to freshly cut branches, Tolkowsky in 1938 concluded that 'in fourth century Italy oranges and lemons were actually grown'.[18]

The first scientific treaty on the citrus tree and its fruits was written in Latin in 1646 by Giovanni Battista Ferrari who titled it *Hesperides, sive, de Malorum Aureorum Cultura et Usu Libri Quatuor* (*Hesperides, or Four Books on the Cultivation and Use of Golden Apples*).[19] The Catholic priest dissected all the variants of known citrus fruits and included in the book 79 impressive botanical illustrations realized as colored engravings by some of Europe's greatest painters of the time, including Poussin, Guido Reni, and Anna Maria Vaiani (Fig. 1). Along with the fruit taxonomy, the book included the history and cultivation methods of the fruits.

Figure 1. Engraving of lamia created by Anna Maria Vaiani for G. B. Ferrari's book *Hesperides, sive, de Malorum Aureorum Cultura et Usu Libri Quatuor* (1646). (*Source*: Public domain, via Wikimedia Commons.)

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Gallesio, a monk traveling in Italy in 1523, reported that he saw large plantations of orange, lemon, and citron trees in Sicily and Calabria, as well as along the river Salo in Liguria.[20] By the late 18th century, lemons and oranges were highly appreciated, even in countries where the citrus tree could not grow due to winter frost. For example, in Germany, Goethe, 5 years before traveling to Sicily in 1787, opened the fourth book of a novel published in 1782 with a famous lyric: 'Kennst du das Land, wo die Zitronen blühn,/Im dunkeln Laub die Goldorangen glühn…?' ('Do you know the land where the lemons grow,/Where oranges grow golden among dark leaves…?').[21]

The citrus tree (and its main fruits such as orange, lemon, bergamot, mandarin, grapefruit, clementine, chinotti and citron) has played a significant societal, economic, and cultural role in the whole Italian peninsula.[20] Since the early 18th century, Sicily, Italy's largest region, has hosted a significant citrus agroindustry that has included plantations and citrus processing companies. Second-tier lemons were already in use in Sicily in the 18th century as source of lemon EO and for producing so-called *agrocotto* (concentrated lemon juice obtained by cooking the fresh juice) and, since the 1850s, also as a source of calcium citrate.[22]

Due to the unique stability of the harvested fruit, first-tier lemons were exported by ship to North Europe and to North America, ensuring 'fabulous profits' for citrus growers in Sicily.[23] Export of citrus fruits to North European countries was carried out also by other countries such as Spain and Portugal. Imported to London from Portugal via the Netherlands, for example, oranges, lemons, and their juices were sold in England as early as the 17th century.[24]

In the 1860s, soils cultivated with lemons in Sicily were the most profitable in Europe. In 1876, the agricultural income granted by a *Citrus* grove was around 2500 liras per hectare, whereas Sicily's average land rent was 40–41 liras.[25] The 'fabulous profits' mentioned above lasted until huge regions in Florida, Brazil, and Argentina became large producers and exporters of citrus fruits. The profitable production of EOs and citric acid from lemon juice, however, continued to flourish in Sicily until the late 1920s. Subsequently, the introduction of the sugar fermentation route first in 1919 by *Penicillium* mold in Belgium (and since 1923 by *Aspergillus niger* in New York),[6] quickly outcompeted the lemon-based citric acid production in Sicily.

An increase in productivity from 0.13 to 0.197 t/ha in the two decades between 1996 and 2007[25] led to an increase in the production of oranges in Sicily by 40% (from 840 000 to 1.18 million tonnes). In the same period, the price paid to farmers remained low, varying between €0.286/kg and €0.34/kg for high-quality blond oranges (chiefly sold for domestic consumption). Along with the spread of pathologies such as mal secco disease,[26] prolonged low fruit prices explain the reduction in the overall cultivated land from 106 644 ha in 2000 to the current 88 000 ha.

Lemon production has experienced a similar situation. It amounted to less than 300 000 t with the price paid to farmers being on average to €0.50/kg.[27] The situation started to change in the early 2010s, however, first with the emergence of e-commerce directly connecting fruit producers with consumers, and then with the coronavirus 2019 (COVID-19) emergency. The latter enabled the rediscovery of the beneficial health properties of lemon, driving the price to €0.8–0.9/kg in the second half of 2020.[28] The trend continued and, in early October 2023, organically grown Sicilian Verdelli lemons (a green-colored lemon with a unique flavor) were sold online by a Sicilian company at €2.40/kg.[29]

In 2023, finally, the price of orange juice reached historic high values (Fig. 2) – over 270% higher than in 2020. In the first 9 months of 2023 the price of orange juice increased by 70%, reaching \$3.66/lb by early October 2023.[30]

Sicily hosts about 20 companies processing citrus fruits, extracting (and concentrating) the juice after recovering the EOs (generally via cold pressing).[31] Their revenues exceed €300 million. For example, one company producing chiefly EOs and commercializing its own natural citrus fruit juices in 2021 had revenues exceeding €27 million.[32]

Another, producing both citrus fruit juices and EOs in the same year, had revenues exceeding €25 million.[33] Demand for orange (and lemon) juice is high, whereas the price of orange and lemon EOs has increased by nearly one order of magnitude in the last 15 years (2007–2022) with the price of orange oil increasing from \$1.4/kg in 2007 to \$9.9/kg in December 2022.[34]

After decades in which highly perishable WOP was disposed of by companies through landfilling, a significant and increasing fraction of Sicily's 340 000 t WOP annual output is mixed with other biowaste and used as anaerobic digestion feedstock to produce biogas and organic fertilizer.[35]

A single biodigester plant located in central Sicily currently processes 30 000 t of WOP yearly. The latter, following mixing with other vegetable and animal biowaste, undergoes anaerobic fermentation affording a 500 m^3/h biomethane production rate.[36] Other citrus processing companies dry WOP to produce pectin raw material and use wastewater from the juice production process as raw material for biogas production, allowing the soluble sugars present in wastewater to undergo fermentation to produce bioethanol.[37]

Finally, other companies supply the WOP for ruminant feeding, which in principle is a way to reduce the impact of feedstuff transport and reuse a readily available feedstock; but

Figure 2. Orange juice price between late 2012 and late 2023. (*Source*: Markets Insider, 2023.)

in practice this is limited by the bitter flavor of WOP, which makes this substrate not very attractive to ruminants, and by the low nutritional value of dried waste peel, requiring protein integration to compensate for its poor nutritional value.[38]

Other citrus processing companies supply fresh lemon peel to a company managing a pectin production plant located along Sicily's northern coast. In 2022 this plant, which is part of Sicily's citrus agroindustry, had revenues exceeding €20 million.[39]

Bioeconomy and green chemistry

The above suggests that the citrus economy in Sicily is a highly dynamic agro-industrial sector comprising a few (about 20) wealthy citrus fruit-processing companies alongside over 40 000 fruit-growing companies, which, after decades of low citrus fruit prices, increasingly benefit from the Internet, which enables them to sell their fruits directly to customers at prices that are three to five times higher than the €0.3–0.4/kg price paid by large buyers.

The fruit processing companies, however, benefit increasingly by saving on WOP disposal costs as they supply their main by-product as feedstock for anaerobic digesters, thereby disposing of the €100/t unit cost (and not the €30/t rate often mentioned in the literature)[4] needed to landfill WOP in authorized landfills.[40]

This dynamic situation has also been reflected in the bioeconomic research carried out in Sicily in the last decade (2012–2022) by researchers at universities and research institutes based in Sicily, on the valorization of citrus processing waste. Selected achievements include the discovery of microwave-assisted[41] and hydrodynamic cavitation-assisted[42] (HC-assisted) extraction processes carried out in water alone to obtain pectin, cellulose, biophenols and EOs; the optimization of biogas production from WOP,[36] and from industrial wastewater;[37] the use of WOP as source of cellulose (extracted with hydrogen peroxide under basic conditions) to make textile fibers;[43] the use of WOP as soil biofertilizer,[44] and to produce thermal insulating panels for the construction industry.[45]

Clearly, in a island with high solar irradiation levels like Sicily, there is room for the utilization of value-adding conversion technologies applied to WOP using solar energy such as that developed by Chemat, Mandi, and co-workers, again employing water as the only extraction medium.[46]

With regard to the practical feasibility of bioproductions based on citrus processing waste, research papers often omit to consider the highly perishable nature of this biowaste, which contains plentiful sugars, water, and residual air that are quickly used by acetic acid bacteria and yeasts to produce acetic acid and ethanol.[47] Reviewing progress after more than a decade of fundamental and applied bioeconomy research efforts it is therefore instructive to learn from failures as well as from successful applications.

The only two uses of WOP that are significant from a practical viewpoint as alternatives to landfilling in Sicily are biogas production in incentivized biogas anaerobic digesters, and as raw material for pectin production. This is not surprising. Citrus-processing companies earning large and increasing revenues from conventional citrus juice and EO production do not have any financial difficulty in bearing biowaste disposal costs. Hence, one might ask when (and if) such companies might wish to invest in new technology to produce new bioproducts from the main by-product of their productions. In this case, one can learn from successful bioproductions from abroad.

In agreement with recent guidelines for bioeconomy companies, which indicate that the production of low-volume, high-margin bioproducts should be targeted,[15] a company based in Great Britain has commercialized, for over 20 years, a registered natural biocide derived from valorized orange pith and peel, which, combined with natural flavonoids and organic acids, acts as a powerful antimicrobial.[48] Another company in the same country manufactures an antimicrobial agent based on citrus flavonoids extracted from immature bitter orange (*Citrus aurantium*) combined with small amounts of malic and citric acids. Patented in the early 2000s, the core product formula is used commercially in many applications including food shelf-life extension, oral care,[49] and surface and hand decontamination.

Citrus flavonoids are highly valued commercial compounds. Their commercial value is well known to citrus processing companies based in Sicily. For example, one such company manufactures and commercializes food-grade commercial citrus peel powder extracts, rich in highly bioactive hesperidin, recovered from waste orange peel after juice extraction.[50]

One tonne of WOP contains about 400 g of hesperidin, a highly bioactive flavanone that is currently commercialized[51] at €1560/kg as a main ingredient of food supplements that contribute to the reduction of cardiovascular risk.[52]

Similarly, the high economic value of substances such as citrus flavonoids, but also of pectin and of microcrystalline cellulose, is known to citrus processing companies. The main reason why most citrus processing companies worldwide have not adopted these bioproduction methods, we argue, lies in the high capital and operational costs of conventional bioproduction routes. The case of pectin, in this respect, is emblematic. Applications of this uniquely versatile food hydrocolloid are expanding rapidly beyond its traditional use as gelling and texturizing agent in the food and beverage industries, with demand (and price) growing at substantial rate for at least a decade.[14] Yet industrial production, using a hot aqueous solution acidified with mineral acid followed by precipitation with alcohol, also creates large amounts of wastewater. This requires an expensive plant, including a solvent recovery unit, in which the costly pectin extraction and purification processes are carried out with substantial energy consumption.

What is needed to change this situation, therefore, are new green chemistry extraction and purification technologies that, using milder reactions and avoiding the use of chemicals such as mineral acid, base, and organic solvents, do not generate biowaste requiring expensive processing.

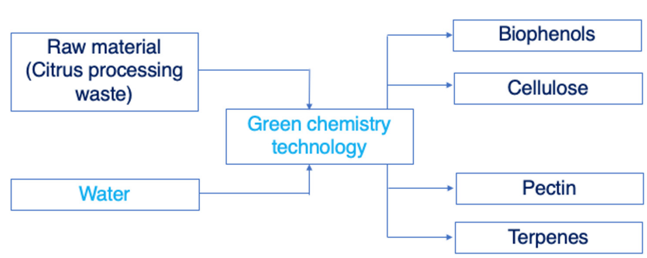
Similar new extraction and isolation technologies allowing new bioproductions in continuous, modular plants affording little or no waste, preferably using water only (i.e., no chemicals and organic solvent – Scheme 1) exist and include hydrodynamic[42] or acoustic[53] cavitation (AC), followed by membrane separation.[54]

The use of membrane technology to purify and isolate pectin by combining ultrafiltration with diafiltration, for instance, is far better than pectin precipitation with ethanol or isopropyl alcohol in terms of concentration, galacturonic acid content, and yield.[54] It is therefore not surprising to learn that the first plants using such new technologies have recently commenced operation.[55]

Indeed, the rapidly expanding pectin market and applications is creating room for new production methods carried out in new production plants using green and modular chemistry technology to meet changing market demands in a flexible way.

It is instructive that, after decades in which the only use of membrane technology in the citrus industry has been for juice clarification, in the second half of the 2010s membranes started to be used in some citrus-processing companies to recover flavonoids and other phenolic compounds present in wastewater via environment-friendly membrane-based technologies for separation, fractionation, and concentration.[56]

It is also revealing that large citrus industry companies have recently established new companies aimed at commercializing natural food ingredients derived from waste orange peel. One such company, for example, commercializes a new fiber 'with natural orange taste and neutral flavour'[57]



Scheme 1. Green chemistry technology affording integral conversion of citrus processing waste into valued bioproducts.

suitable for use as a low-calorie texturizer and source of dietary fiber in numerous foods (baby food, bakery, confectionary, desserts, toppings and fillings, jams and jellies, frozen meals, sauces, creams, etc.) and beverages – namely nearly all of the industry's market segments in which citrus pectin is employed.[14]

Outlook and conclusions

Analysis of the citrus economy in Sicily at the dawn of the bioeconomy era reveals numerous findings that are relevant for most citrus-growing countries.

The first relevant finding is that, with the Internet directly connecting farmers and consumers, a number of Sicilian citrus producers no longer sell their valued oranges at €0.3–0.4/kg or their lemons at €0.5/kg, but instead sell them at prices that are three to five times higher.

A second interesting finding is that, because market prices have reached historically high levels, Sicily's citrus processing companies that successfully produce citrus juices and essential oils[58] earn large and increasing revenues.

Third, anaerobic digestion of waste orange peel has clearly emerged as the main use of citrus processing waste as an alternative to landfilling, with citrus-processing companies disposing of their waste citrus peel by making it available for free to biodigester plants (thus eliminating biowaste disposal costs).

Fourth, showing evidence of market awareness of the high economic value of bioproducts present in citrus fruits, selected citrus processing companies already extract valued flavonoids such as hesperidin from orange peel and commercialize them.

In conclusion – in contrast with what happened when the emergence of new production technology killed Sicily's citric acid industry based on lemon juice[6] – when new green chemistry bioproduction technologies such as cavitation (hydrodynamic and acoustic), microwave-assisted extraction, and advanced fermentation to produce valued chemicals such as xanthan gum are commercialized, citrus processing companies will start using fresh waste citrus peel to produce and commercialize valued bioproducts. The market demand for these bioproducts is so large that, for many years to come, increasing supply will not be able to meet demand.

Waste orange peel, for instance, is an excellent source of microcrystalline cellulose (MCC), a partially depolymerized non-fibrous crystalline powder composed of porous particles.[59,60] Generally derived from wood, MCC is used in numerous drug formulations as the excipient that has 'revolutionized tableting.'[61] Microcrystalline cellulose is also used by the cosmetic and food industries as an emulsifier, stabilizer, anti-caking agent, texture modifier, stabilizer, fat substitute, and suspending agent, leading to a $1 billion per year market growing at an annual rate of ~7%.[62] The analogous citrus processing waste of lemon and grapefruit subjected to hydrodynamic cavitation affords, along with highly water-soluble, low methyl-esterified and highly bioactive pectin 'IntegroPectin,'[62] a micronized cellulose dubbed 'CytroCell,' which is similar to nanocellulose.[63]

By collecting and providing updated information, that previously was not updated, which was scattered, and often poorly known even to bioeconomy practitioners, this study aims to contribute further to this progress.

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Conflict of interest

The authors declare no conflict of interest.

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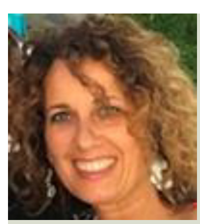

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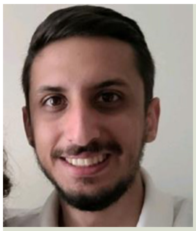

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