LIFE CYCLE ASSESSMENT FOR VALORIZATION OF ORANGE AND APPLE JUICE RESIDUES

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ABSTRACT

LIFE CYCLE ASSESSMENT FOR VALORIZATION OF ORANGE AND APPLE JUICE RESIDUES

Healthy diet trends increased the demand for natural and nutrient beverages, including fruit juices. In the ever-growing fruit juice market, orange and apple juice are the most popular flavors. Nearly half of the cultivated fruits are processed into juice, creating a great amount of residue. The current waste management techniques for these residues are landfilling or composting. Instead, they can be valorized to produce value-added products within the circular economy model. As the resources are finite, transitioning to a circular economy model is crucial for a sustainable future. Novel technologies have been emerging to support a circular and biobased economy to utilize agricultural waste. In this study, life cycle assessment (LCA) methodology was applied for orange juice residues to produce d-limonene and pectin; and for apple juice residues to produce succinic acid and xylitol. D-limonene was extracted by cold-pressing and solvent-free microwave extraction and pectin was extracted by acid hydrolysis. Waste apple pomace was used to produce succinic acid and xylitol by fermentation. The valorization routes were compared with landfilling, showing all of them performed better environmentally except succinic acid. 3 future scenarios were created based on the decarbonization of electricity and agriculture for climate goals. Results showed that the environmental impacts were caused by intensive electricity use in production and agricultural production, so valorization routes would perform better in future scenarios with decarbonization in electricity and agriculture.

ÖZET

PORTAKAL VE ELMA SUYU POSALARININ DEĞERLENDİRİLMESİ İÇİN YAŞAM DÖNGÜSÜ DEĞERLENDİRMESİ

Sağlıklı beslenmenin popülerleşmesi ile meyve suları dahil olmak üzere doğal ve besleyici içeceklere olan talep artmıştır. Günden güne büyüyen meyve suyu sektöründe portakal ve elma suyu en popüler aromalardır. Üretilen meyvelerin neredeyse yarısı meyve suyu olarak işlenmekte ve büyük oranda posa atığı oluşmaktadır. Bu atıkların yönetiminde düzenli depolama ve kompostlama kullanılmaktadır. Bunun yerine, döngüsel ekonomi modeli çerçevesinde bu artıklar katma-değerli ürünler üretmek için kullanılabilir. Kaynaklar sınırlı olduğundan sürdürülebilir bir gelecek için döngüsel ekonomiye geçiş kritiktir. Döngüsel ve biyobazlı ekonomiyi desteklemek adına tarım artıklarını kullanmak için yenilikçi teknolojiler geliştirilmektedir. Bu çalışmada, portakal suyu posasının d-limonen ve pektin üretiminde kullanılması ve elma suyu posasının süksinik asit ve ksilitol üretiminde kullanılması için yaşam döngüsü değerlendirmesi (YDD) metodolojisi uygulanmıştır. D-limonen için soğuk presleme ve solventsiz mikrodalga ekstraksiyonu; pektin için asit hidrolizi kullanılmıştır. Süksinik asit ve ksilitol için fermentasyon kullanılmıştır. Bu değerlendirme yöntemleri düzenli depolama ile karşılaştırılmış ve süksinik asit dışında tüm yöntemler daha iyi çevresel performans göstermiştir. İklim hedefleri çerçevesinde elektrik ve tarımsal üretimin karbonsuzlaştırılması baz alınarak 3 gelecek senaryosu oluşturulmuştur. YDD sonuçları çevresel etkilerin yoğun elektrik kullanımı ve tarımsal üretimden kaynaklandığını, dolayısıyla elektrik ve tarımda karbonsuzlaşma ile gelecek senaryolarda değerlendirme yöntemlerinin daha iyi çevresel performans göstereceğini ortaya koymuştur.

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LIST OF SYMBOLS/ABBREVIATIONS

ADP fossil	Abiotic Depletion Potential Fossil Resources
AP	Acidification Potential
CAJ	Concentrated Apple Juice
CBP	Consolidated Bioprocessing
EP	Eutrophication Potential
FC	From Concentrate
FCOJ	From Concentrate Orange Juice
GHG	Greenhouse Gas
GWP	Global Warming Potential
IEA	International Energy Agency
LCA	Life Cycle Assessment
MJ	Megajoule
NFC	Not From Concentrate
NREL	National Renewable Energy Laboratory
NZE	Net Zero Emissions
OPW	Orange Peel Waste
PV	Photovoltaic
R&D	Research and Development
RFC	Reconstituted From Concentrate
SFME	Solvent-Free Microwave Extraction
SHF	Separate Hydrolysis And Fermentation
SSCF	Simultaneous Saccharification And Co-Fermentation
SSF	Simultaneous Saccharification And Fermentation
TOA	Tri-n-octylamine
TCA	Tricarboxylic Acid
v/v	Volume Per Volume
Vvm	Vessel Volumes Per Minute
w/w	Weight By Weight
WAP	Waste Apple Pomace
WWTP	Wastewater Treatment Plant

1. INTRODUCTION

In the last decade, the food processing industry has grown remarkably with developing technologies and increasing population. The increasing trend in health and wellness also increased the demand for natural and nutrient-rich beverages, particularly fruit juices since they are perceived as a good source of vitamins and antioxidants (Imarc Group, n.d). The fruit juice market was valued at 146.23 billion USD in 2022 and is projected to reach 196.49 billion USD with an expected 4.31% growth between 2023-2029 (Data Market Research, 2022). The market is segmented according to flavors and orange juice has the biggest share, followed by apple juice (Agri Exchange Portal, n.d.). In the Turkish fruit juice market, there are 6 main fruits processed in the fruit juice sector: Apple, peach, apricot, cherry, orange, and pomegranate. (Meyve Suyu Endüstrisi Derneği, 2011). According to FAO data, Türkiye is ranked as the 10th largest agricultural country in the world as of 2016, with agricultural production for food and beverages reaching 64 billion USD (Güney Ege Kalkınma Ajansı, 2020).

Nearly half of the cultivated fruits are processed into juice. 20-80% of the whole fruit weight ends up as residue after the fruit processing (Kandemir et al., 2022). 25-30% of the waste generated from fruit and vegetable processing is not utilized, creating great amounts of waste (Sagar et al., 2018). Since the resources are finite, adapting to a circular economy model is essential for a sustainable future. Valorization is a key concept for circular economy transition since it aims to reduce waste and conserve resources while promoting sustainable development. The EU Commission describes biodegradable waste as “any waste that is capable of undergoing anaerobic or aerobic decomposition, such as food and garden waste, and paper and paperboard” and set targets to reduce the landfilling of biodegradable waste through recycling, composting, biogas production or materials or energy recovery in this sense (Directive 1999/31/EC).

In line with this approach, fruit waste should be considered as a renewable, secondary raw material to transition into a circular economy. Up to half of the whole fruit weight of oranges and up to 25% of the fresh fruit weight of apples end up as residue after juicing (Teigiserova et al., 2022; Waldbauer et al., 2017). With their dominance in the fruit juice industry, orange and apple juice residues have a great potential for valorization. Through the valorization routes, value-added products can be extracted and produced from the fruit juice residues. Waste apple pomace (WAP) and orange peel waste (OPW) as juicing residues hold great potential for valorization since they

have a high production rate globally and in Türkiye and they have rich chemical composition. Furthermore, there is an increasing research interest in bio-based chemicals, hence new technologies are developed. Two major value-added products that can be extracted from orange peel waste are d-limonene and pectin (John et al., 2017). The high sugar content of waste apple pomace allows it to be utilized for fermentation to produce various value-added chemicals.

This study aimed to analyze the potential environmental impacts of producing value-added products from fruit juice residues by life cycle assessment. Orange juice and apple juice residues were selected due to their high production rate and high valorization potential. Within the scope of this study, LCA methodology was applied for d-limonene and pectin production from OPW, and succinic acid and xylitol production from WAP. These chemicals were selected according to their market potential and the availability of their industrial production. The valorization routes were compared with landfilling, which is the current waste management scenario for fruit juice residues. Decarbonization scenarios for 2035 and 2050 were applied for agricultural production and electricity production. Environmental hotspots were spotted through LCA.

2. LITERATURE REVIEW

2.1. Orange Juice: Production and Waste Management

In the current era of wellness-focused living, the consumption of fruit juices has emerged as a notable and highly desired element of a healthy lifestyle. Consumers have become more aware of the nutritional benefits of fruit juices which became a driving force for the growth of the fruit juice market; the global juice market grew to 141 billion USD with a production volume of 67.6 billion liters in 2022 (Esturo et al., 2023). Orange juice is the top choice among consumers in the 100% juice category, capturing 43.8% of the market, and it maintains its lead in the nectar category with an 18.9% consumption share (Neves et al., 2020). Orange juice is among the top-produced juices in Türkiye, produced under 10.32.12 NACE code and according to 2020 data, the capacity for orange juice production is 36,054,530 liters in Türkiye. According to the orange cultivation data, Antalya is the leading province by 31% and Adana comes in second place by 22%. Antalya and Adana also have the biggest orange juice production capacity with 6 million liters and 6.5 million liters respectively (Batı Akdeniz Kalkınma Ajansı, 2020).

Orange juice is produced in two different ways; either as a single-strength bulk product in frozen or aseptic form, called not-from concentrate orange juice (NFC), or as a frozen bulk concentrate, called frozen concentrate orange juice (FCOJ) ("Orange Book", 2023). NFC is 100% freshly squeezed orange juice. FCOJ is a concentrated product made by removing water from orange juice and then freezing it, to preserve its freshness ("Orange Book", 2023). In Türkiye, orange juice is produced under the 10.32.12 NACE code with 3 products: 10.32.12.10.00 orange juice, frozen (not concentrated), 10.32.12.20.00 orange juice, not frozen (not concentrated) and 10.32.12.30.00 orange juice, not elsewhere classified (Batı Akdeniz Kalkınma Ajansı, 2020). Two primary kinds of orange juice products are concentrates and ready-to-drink juices. Concentrates are required to be diluted with water before consumption. Ready-to-drink juices are either NFC or reconstituted from concentrate (RFC). Ready-to-drink (RTD) orange juice is at the strength to be consumed, it doesn't need to be diluted before drinking ("Orange Book", 2023). RFC is often reconstituted to its original concentration before marketing (Galaverna & Dall'Asta, 2014).

Orange juice production process starts with receiving the fruits at the plant. Oranges are washed, sorted and separated according to their size and quality to ensure correct feeding into the juicers (Ortiz-Sanchez et al., 2023). There are two types of extraction methods for juice production. In the first approach, fruit is positioned between two metal cups containing sharp metal tubes. When pressure is applied, these tubes pierce the fruit and juice is extracted and collected. In the second approach, oranges are halved, and juice is extracted through moving these halves against rotating reamers. Then various equipment including finishers, hydro cyclones and separators are employed to remove the excessive pulp, seeds and peels (Galaverna & Dall'Asta, 2014). Then a stainless-steel screen is used to filter the juice, which can then be refrigerated or concentrated to create two different types of juice: not from concentrated (NFC) and from concentrated (FC) ("Orange Book", 2023). For NFC orange juice production, the juice is centrifuged and pasteurized to increase shelf life and for FC orange juice the juice goes to the evaporator to remove water then it is frozen for storage (Ortiz-Sanchez et al., 2023). The juicing process is given in the Figure 2.1 below.

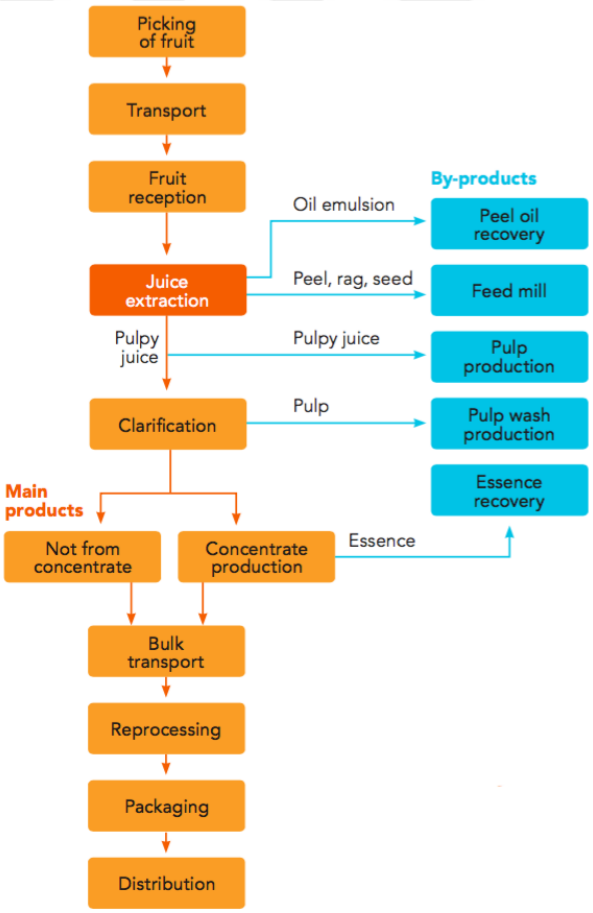

Figure 2.1. Orange juice production process ("Orange Book", 2023).

Nearly half of the fresh orange weight is processed to produce orange juice and the other half, including peel, pulp and seeds, are discarded as residue (Rezzadori et al., 2012). Considering the size of the global orange juice production, a huge amount of waste is created. According to a study from 2019, 55 million tons of orange peel waste is generated annually (Teigiserova et al., 2022). Considering the volume of waste and its properties, careful and immediate waste management is needed. Orange pomace and peels contain a high amount of water and are rich in organic matter, making them susceptible to putrefaction so storage and disposal should be planned accordingly (Li et al., 2023).

Current waste management of OPW mainly includes landfilling and composting (Ortiz-Sanchez et al., 2021). These methods have both environmental and economic disadvantages. First, the EU landfilling directive discourages the landfilling of organic waste and mandates minimization (Negro et al., 2017a). Landfilling also has problems such as the generation of leachate and organic compound emissions (Ortiz-Sanchez et al., 2021). When dumped, high amounts of waste start to accumulate and decompose, leading to uncontrolled methane production and creating a risk to water resources (Ángel Siles López et al., 2010). OPW can be composted and used as a fertilizer, however this has advantages and disadvantages. OPW can enhance soil quality by increasing organic content and water retention, and by providing a rich source of nutrients (de la Torre et al., 2019). However, it is a costly process and the compost might not find a ready market locally (Negro et al., 2017a). Also, composting can lower the pH too much which leads to undesired fermentations (de la Torre et al., 2019). Traditionally peels and pomaces have been used as animal feed. When used as cattle feed beef, quality is enhanced and growth is promoted, and it can reduce fat content while increasing meat content if it is added to poultry diets in moderation (2-3%) (de la Torre et al., 2019). However, there are drawbacks related to its use as animal feed since it needs an energy-intensive dehydration process before use, it's bitter and has low nutritional value (Negro et al., 2017a). Incineration is also applied, but it leads to the generation of GHGs, not being the best environmentally viable option for disposal. These problematic disposal methods led to the development of environmentally viable solutions that not only reduce waste but also produce value-added products (Ángel Siles López et al., 2010).

OPW, as a by-product of orange juice production, gained significant research attention in recent years as one of the most studied agro-industrial residues (Ortiz-Sanchez et al., 2021). Its extensive global production and nutrient-rich composition make it a valuable biomass for further research. OPW contains monosaccharides, polysaccharides, fibers, polymers, fat, and proteins. The

main monosaccharides are glucose, fructose, and sucrose, making nearly 30 – 36 % w/w of dry solid. The main polysaccharides are cellulose, hemicellulose, and pectin; where cellulose makes up 18-20%, hemicellulose 14-16% and pectin 20-22% w/w of dry solid. Lignin is an important structural polymer, making up 5-7% w/w dry solid. OPW contains up to 8% w/w of dry solids and the rest include polyphenols, organic acids and essential oil (0.3 – 0.5 w/w dry solid) (de la Torre et al., 2019). Organic acids include citric acid, malic acid, malonic acid, and oxalic acid but they are very low in quantity, making up nearly 1% (Ángel Siles López et al., 2010).

OPW consists of 60-65% peels and 30-35% pulp and seed (Senit et al., 2019). Peel waste consists of two components: The flavedo, the outer orange-colored layer, which is rich in essential oils, carotenoids, phenols and flavonoids; and the albedo, the inner white layer, which is abundant in pectin, cellulose, hemicellulose, lignin and dietary fibers. Pulp waste is rich in fibers such as cellulose, hemicellulose and lignin, pectin, essential oils, polyphenols, flavonoids, and natural antioxidants, while the seeds are abundant in essential oils and limonoids (Dubey et al., 2023). The fruit composition is given in the Figure 2.2. below

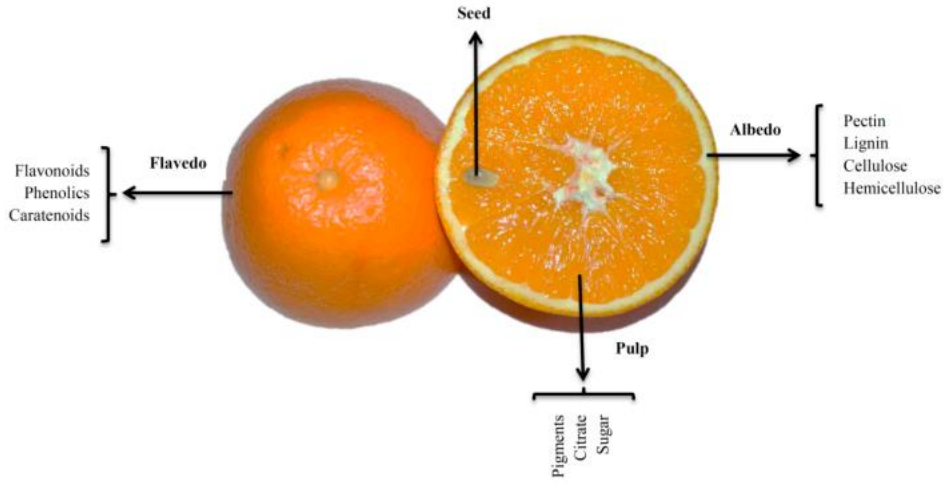

Figure 2.2. Citrus fruit composition (Putnik et al., 2017).

OPW can be used as raw material to produce various products through chemical, thermal, enzymatic or biological conversion, as shown in Table 2.1 below:

Table 2.1. Products from OPW (Adapted from (Ortiz-Sanchez et al., 2023).

Process	Product
Physical	Polyphenolic compounds, Carotenoids, Essential oil, Dietary fiber, Antioxidants, Pectin
Chemical	Ascorbic acid, Ellagic acid, Xanthan, Levunilic acid, Biodiesel, P-Cymene
Biotechnological	Butanol, Lactic acid, Mucic acid, Citric acid, Succinic acid, Ethanol
Thermochemical	Hydrogen, Activated carbon, Bio-oil, Biochar

Knudsen et al. (2011) investigated the environmental hotspots of organic orange juice in small producers in Brazil, processed and imported to Denmark by using LCA approach, which revealed that transport is the main contributor, followed by the farming stage in global warming potential category. Mariana et al. (2021) investigated an environmental analysis for various biorefinery scenarios starting from orange cultivation to valorization routes, revealing that the orange processing into juice has an impact of 1.96 kg CO_2 eq. to climate change category when mass allocation is applied. There was an EU project that investigated the LCA for orange juice as a deliverable, showing that the bottling process is the main contributor to climate change and abiotic resource depletion categories, whereas orange cultivation is the main contributor to the rest of the categories (Doublet et al., 2013).

2.1.1. D-Limonene

D-Limonene is a terpene, commonly found in essential oils derived from citrus fruit skin (Aissou et al., 2017). It is a clear liquid with an orange scent, taking its name from lemon, limonene is present in various citrus fruits and it gives the characteristic smell to them (El-Ishaq et al., n.d.). Limonene makes up to 90% wt. of the essential oil found in the orange and lime peels (Ramos-Ibarra et al., 2021). Commercial extraction of d-limonene began in the 1940s in Florida, USA from the steam evaporator condensate used in citrus molasses production (*D-limonene, 1979*). Orange juice industry is a key source of d-limonene, it is mostly extracted from waste orange peels, which have 3.8% of d-limonene on a dry weight basis (Ciriminna et al., 2014).

D-Limonene is acknowledged as GRAS (Generally Recognized as Safe) by the U.S. Food and Drug Administration and is extensively utilized in flavors and fragrances; also serving as a cleaning/degreasing agent in both industrial and household applications, with increasing attention (Aissou et al., 2017). D-limonene and orange oil/essence oil (containing 95% d-limonene) have

been used as flavoring and fragrance agent in perfume, soap, food and beverages, non-alcoholic beverages, ice cream, sweets, baked goods, gelatins, puddings, and chewing gum for nearly 50 years (*D-limonene*). D-limonene was first promoted as a versatile and environmentally friendly solvent that can substitute fossil-based toxic solvents. Cleaning products for both industrial and household use were the primary market d-limonene, which could effectively replace harmful solvents like toluene, n-hexane, and chlorinated organic solvents in various industrial applications (Ciriminna et al., 2014). Clinically, d-limonene has been used to dissolve gallstones and lighten heartburn while it also has shown chemo-preventive properties against various cancer types (Hardjono et al., 2021). Registered as an insecticide and anti-bacterial, it is one of the first natural pesticides for an environmentally friendly pest control ingredient (Ciriminna et al., 2014). The global market for d-limonene was valued at 473.72 million USD in 2021 (Data Market Research, 2022). The demand rises as it has the potential to substitute hazardous components in solvent blends and the global demand is expected to surpass 65 kilotons by 2023 (John et al., 2017).

At present, the primary source of limonene is citrus peels obtained from fruit processing in citrus juice industries, usually through cold-processing methods including centrifugal separation or steam distillation (Becerra & Villa, 2020). The oil sacs are located at the flavedo and several traditional and novel extraction methods are used for the extraction. Various conventional extraction methods such as cold pressing, hydro-distillation, steam distillation and solid-liquid extraction with organic solvents, are employed for d-limonene extraction from citrus waste (Negro et al., 2017b; Siddiqui et al., 2022). Various alternative organic solvents are under research to find a sustainable, cost-effective, environmentally friendly solvent to achieve the highest possible yield. Hexane is the most used solvent with a 99% recovery yield; however, it is a fossil-based chemical that poses environmental and health hazards. Bio-based solvents such as cyclopentyl methyl ether and 2-methyl-tetrahydrofuran (2-MeTHF) and natural deep eutectic solvents are researched as alternatives (Siddiqui et al., 2022). The conventional extraction methods exhibit several drawbacks such as extended extraction time, loss of volatile compounds, thermal degradation, low extraction yields and high energy and solvent use (Negro et al., 2017b). Novel technologies gained attention to address these drawbacks. These methods include ultrasound-assisted extraction, microwave extraction and controlled pressure drop, among them microwave-assisted extraction (SFME) is the most promising solution since it is a simple process that does not utilize water or organic solvents (Auta et al., 2018). Supercritical fluid extraction and enzyme-assisted hydro distillation (EAHD) are also researched as alternative methods (Siddiqui et al., 2022).

Valorization of citrus peels as well as orange peel waste from the juicing process have gained attention in the literature and related environmental impacts and life cycle assessments are available for various processes. Available research mostly adapted an integrated approach for biorefinery, combining d-limonene production with other products. Mariana et al. (2021) evaluated four scenarios for orange peel waste, through life cycle assessment. The scenarios include biorefineries with different complexities that combined the production of essential oil, pectin, biogas, acetone, butanol and ethanol and the base scenario is landfilling. A similar integrated process is seen in Pourbafrani et al. (2013)'s study for life cycle greenhouse gas impacts to produce ethanol, biomethane and limonene from citrus waste, where two scenarios for small and large biorefineries are assessed. Teigiserova et al. (2022) did a life cycle assessment for a biorefinery that utilizes orange peel waste, to produce essential oil, citric acid and mycelium. Da Costa et al. (2022) analyzed the environmental performance of orange waste to produce pectin and essential oil.

2.1.2. Pectin

Pectin is a natural heterogeneous polysaccharide, primarily composed of galacturonic acid (GalA) monomers that are commonly found in the cell walls and middle lamella structures of all higher plants (Wang et al., 2014). French pharmacist and chemist Louis Nicolas Vauquelin identified pectin's chemical composition in 1790 and later another French pharmacist and chemist Henri Braconnot discovered the gellification properties and named it "pectin" (Canteri-Schemin et al., 2005). While pectin is found in many plant tissues, the pectin used in commercial applications is primarily sourced from citrus peel or apple pomace, since they have higher quality and provide enough for cost-effective production (Casa et al., 2021). Commercial applications require a minimum purity of 65% GalA. The degree of esterification (DE) is influenced by the methyl ester groups within the GalA backbone and according to DE, pectin is categorized as either high DE (>50% methyl esters) or low DE (<50% methyl esters) which also influences its rheological characteristics (Nadar et al., 2022).

Pectin has thickening, gelling and stabilizing properties thus used commonly in the food, cosmetics, medical, chemical, textile, and pharmaceutical sectors (Wang et al., 2014). In food products; it is used as a gelling agent in jellies, as a texture-stabilizing agent in fruit juices and acidified milk, as an antioxidant in spreads and ice cream, and to replace fat in low-fat foods (Mahmoud et al., 2022). In pharmaceuticals, pectin is used in drug encapsulation, developing medicines for ulcers and intestinal issues, and as a potential raw material for creating edible films

and coatings (Casa et al., 2021). It has numerous health effects; it has a beneficial impact on blood cholesterol levels and serum glucose levels and serves as a natural preventive measure against toxic cation poisoning, effectively eliminating lead and mercury from the gastrointestinal and respiratory systems (Thakur et al., 1997; Venkatanagaraju et al., 2020). With its extensive use, the pectin market is expected to reach 0.96 billion USD in 2023, with Europe having the biggest market share (Mordor Intelligence, n.d).

Pectin is abundant in the cell walls of most plants thus it has been isolated from a wide range of plants such as apple, citrus peel, carrots, sugar beet pulp, sunflower heads, papaya and oranges (Venkatanagaraju et al., 2020). Apple pomace and citrus peel have pectin content of nearly 10-15% and 20-30% respectively on a dry matter basis, while sunflower head residues and sugar beet contain about 10-20% pectin on a dry weight basis (Chandel et al., 2022). Pectin is commercially produced from citrus peels such as lemon, lime and oranges by 85.5, the remaining part comes from apple pomace (14%) and sugar beet pulp (0.5%) (Nadar et al., 2022). Pectin is isolated from the plant material by various methods. It is extracted via single methods such as acid, alkali and enzyme extraction or combined with other methods such as microwave-assisted or ultrasound-assisted extraction to improve efficiency (Mahmoud et al., 2022). The industrial production of pectin is done in acidic conditions at high temperatures but considering the environmental impact of acidic wastewater and high temperatures; alternative extraction methods to use ultrasound, enzymatic, microwave, and subcritical water have gained attention (Wang et al., 2014). These novel techniques are explored for their lower energy consumption, reduced temperature, less reagent use and shorter process time (Benassi et al., 2021).

Enzyme-assisted extraction utilizes enzymes with high specificity to release protopectin from cell walls, without using corrosive acids; aiming to obtain superior quality pectin with increased yield under mild temperatures. Despite advantages such as reduced energy consumption, high-quality pectin, and increased yields; there are potential drawbacks including additional costs of buffers, economic limitations due to the expensive enzymes and concerns about the energy-intensive production process (Nadar et al., 2022). Ultrasound-assisted extraction utilizes mechanical vibrations of ultrasound waves, leading to cell disruption, improved solvent entrance and intensified mass transfer in plant tissue. The advantages of ultrasound over conventional methods include low energy consumption, reduced treatment time, decreased solvent use, and increased yield and efficiency (Marić et al., 2018). Microwave-assisted extraction utilizes a microwave field to heat a dielectric material like water, resulting in rapid and efficient heating due to the nature of water

molecules. The addition of mineral or organic acids for pH adjustment, along with microwave heating significantly reduces extraction time compared to conventional heating methods. Despite its lowered extraction time, the use of microwave heating might lead to higher electricity use (Nadar et al., 2022). Subcritical water extraction utilizes water at subcritical conditions and due to its unique conditions at this state, it is effective for the hydrolysis of lignocellulosic material, without the need for adding acids (Wang et al., 2014). Although it has the benefit of using only water and eliminating acid use; the use of subcritical water can be costly. These green novel technologies hold the potential to decrease the environmental impacts of pectin production, however, there is room for technical development for them to be considered commercially viable processes.

The conventional pectin extraction process can be summarized in three steps: Pretreatment, pectin extraction and purification. Washing, blanching or simple drying are used as pretreatment to deactivate enzymes or microbes to prevent pectin degradation and ensure the production of stable raw material (Nadar et al., 2022). The pectin extraction is usually done under acidic conditions. In the commercial process, raw material is treated with a hot, diluted mineral acid at a pH of approximately 2, involving the use of mineral acids such as nitric acid, hydrochloric acid, sulfuric acid, phosphoric acid and citric acid (Chandel et al., 2022; Sriamornsak, 2003). Downstream processing is crucial to obtain high quality and yield. Purification steps start with filtration, then pectin is washed and precipitated by alcohol, usually ethanol. Then pectin is dried and ground (Casa et al., 2021). There are environmental drawbacks related to this process; the main drawback of using mineral acids is their toxicity and generation of harmful waste which leads to the investigation of using organic acids such as citric acid, tartaric acid and maleic acid as an alternative (Casa et al., 2021). The pectin yield is highly dependent on extraction conditions, including temperature, extraction duration, pH and raw material (Chandel et al., 2022). The process conditions may vary according to raw material, but usually the process requires heating up to 80 – 100 °C, extraction time of around 30 – 180 minutes and atmospheric pressure (Benassi et al., 2021; Casa et al., 2021).

Various research focus on pectin extraction from different biomass, however, only selected few studies focus on environmental impacts and/or LCA for such production processes. Scherhaufer et al. (2020) researched environmental assessment of the production of pectin from apple pomace, along with other food side flows. According to this study, pectin from apple pomace has a GHG emission of 106 kg CO_2e/t apple pomace, which is higher than the average emissions of pectin produced in the EU. This is mainly caused by the heating that is used for drying apple pomace and

the pectin production process (da Costa et al., 2022). Garcia-Garcia et al. (2019) did the first LCA for a pilot-scale pectin production for a microwave-assisted extraction process without the use of acid. In the study, microwave-assisted extraction is compared with conventional acid extraction, resulting in approximately 75% reduction for all environmental impact categories. Dávila et al. (2015) ran a techno-economic and environmental assessment for an integrated production of p-cymene and pectin from orange peel and compared 2 scenarios with and without electricity generated through the gasification of remaining process residues. Both techno-economic and environmental assessments show that the scenario without electricity generation is more attractive. Integrated processes and biorefinery approaches are attractive topics for researchers. Shinde et al. (2020) designed a biorefinery process for pomegranate peel waste to extract ellagic acid, lignin and pectin. According to the LCA results, the pectin extraction stage has the highest contribution to all environmental indicators, caused by the indirect emissions from the production of ethanol. Orange waste is researched to be valorized in integrated processes by da Costa et al. (2022) for pectin and essential oil production, and by Mariana et al. (2021) to analyze biorefineries with different complexities. LCA study by da Costa et al. (2022) revealed that pectin extraction leads to most of the environmental impacts, because of the high energy consumption and materials, especially in the acid hydrolysis stage. Mariana et al. (2021) compared various biorefinery scenarios (including essential oil, pectin, hesperidin, and biogas production) through LCA, revealing biorefineries have a better environmental performance than OPW disposal to landfills. When biorefineries are compared, low-complexity biorefineries have a lower environmental impact as they have a lower energy demand and waste stream, however, energy and water demand increase as the complexity increases, pointing out that the environmental impact is directly proportional to the complexity of biorefinery.

2.2. Apple Juice: Production and Waste Management

Globally apple juice holds the second position following orange juice, with China leading in both the production and export of concentrated apple juice (CAJ) (Cheng et al., 2022). In Türkiye, apple juice and orange juice hold the first two places with 32.8% and 32.4% respectively, according to fruit juice and nectars by flavors (GlobalData Plc, 2018). Apple is a trendy fruit that can be cultivated in regions with temperate climates. Most of the apples are consumed fresh and around 13% are used in apple juice manufacture (Vukušić et al., 2021).

At present, there is a diverse range of apple juices with various processing methods available in the market, with clear juices reconstituted from concentrate (FC) being the most common one (Włodarska et al., 2017). On the other hand, technologies are progressing to produce cloudy juices as a healthier option for clear juices; since compounds that provide health benefits, such as phenolics and pectin, are enzymatically degraded during clarified juice production (Massini et al., 2018). The production of apple juice involves multiple stages, including preparation, milling or crushing, pressing, clarification, filtration, pasteurization, concentration, the addition of food additives, and packaging; according to the juice type, additional steps can be added such as mash and juice enzymation for clear juices (Massini et al., 2018). The difference between the production of clear apple juice and cloudy apple juice comes from enzymatic treatment, filtration and homogenization. Apples are first washed to remove any dirt and debris, then sorted by size and quality to remove any damaged or diseased fruits. Then, the apples are ground into a pulp, which is then pressed to extract the juice (Vukušić et al., 2021). To produce clear juice, pectinolytic and/or cellulolytic enzymes are added to increase juice yield and break down pectin since it gives the apple juice cloudy appearance. Subsequently, clarification and filtration steps are applied to remove any remaining pulp or sediments (Massini et al., 2018). Enzymes are not applied to cloudy apple juice production, rather a homogenization step can be applied to create a uniform product. The final step is pasteurization, where the juice is heated to a specific temperature to eliminate any potentially harmful microorganisms. If the product is concentrated, water is removed via evaporation (Vukušić et al., 2021). The whole process is given in Figure 2.3. below.

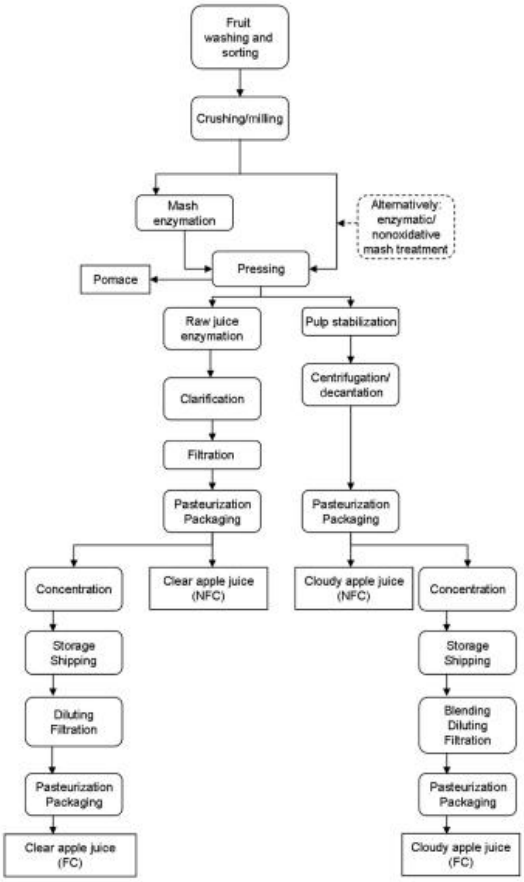

Figure 2.3. Apple juice production process (Neitsch et al., 2011).

Apple pomace is generated as waste after apple juice and cider production and can account for up to 25% of the fresh fruit's weight (Waldbauer et al., 2017). Considering the popularity and production volume of apple juice, apple pomace is generated in great amounts globally. Due to its high biological oxygen demand, disposal of apple pomace poses a pollution risk, also its high water and high fermentable sugar content make it prone to decay (Vukušić et al., 2021). Currently, it is managed through traditional methods like landfilling, incineration, composting, low-quality animal feed and land spreading. However, such disposal can lead to negative environmental impacts, including the generation of greenhouse gases, the risk of secondary pollution with foul odors and groundwater contamination (Dhillon et al., 2013). Also, apple pomace is considered a low-value feed due to its low protein and mineral contents, along with poor digestibility because of its high lignin/cellulose ratio (Awasthi et al., 2021).

The economic and environmental burden of traditional waste management techniques leads to the search for alternatives. This necessity, combined with the rich composition of apple pomace sparked interest in the research of different valorization processes. One of the most important

current uses of apple pomace is to produce pectin, along with citrus peel apple pomace is the main source of commercially available pectin (Marić et al., 2018). Thanks to its rich fiber and polyphenol content, finely ground apple pomace is utilized in the production of food items such as bakery products or crackers to fortify its nutritional value (Barreira et al., 2019). However, it is used in low amounts in order not to change the color, texture and taste, so it has limited use as a food fortification item. Thus, the need and search for utilization maintains for the great amount of available apple pomace.

Apple pomace consists of skin and flesh (95%), seeds (2-4%) and stems (1%) (Vukušić et al., 2021). While the content of apple pomace can differ according to the cultivar, they all are a nutritionally rich source, abundant in phytochemicals, and a significant amount of carbohydrates, alongside smaller proportions of proteins, vitamins, and minerals (Lyu et al., 2020). It is rich in non-starch polysaccharides, including 35-60% dietary fiber, along with high levels of insoluble fiber (36.5%) and soluble fiber (14.6%). Dietary fibers include pectin (5.5–11.7%), cellulose (7.2 - 43.6%), hemicelluloses (4.26–24.40%), lignin (15.30 - 23.50%), and gums. Cellulose, pectin, and lignin constitute the water-insoluble fibers while galacturonic acid and hemicelluloses are water-soluble (Waldbauer et al., 2017). It also has simple sugars such as glucose (22.7%), fructose (23.6%) and galactose (6-15%) (Lyu et al., 2020).

Apple pomace is abundant in soluble carbohydrates like simple sugars (fructose, glucose, sucrose) and polysaccharides (cellulose, hemicellulose, pectin), offering a significant potential for bioconversion (Parmar & Rupasinghe, 2013). Over the past three decades, its potential has been researched as a substrate for microbial growth to produce various value-added products such as organic acids, enzymes, single-cell proteins, low alcoholic drinks, ethanol, biogas, pigments and baker's yeast. (Vukušić et al., 2021). There are studies to produce bioethanol, hydrogen and methane from apple pomace through anaerobic fermentation and additional research has focused on producing gas products via slow pyrolysis of apple pomace to be used as a precursor for hydrogen production (Gołębiewska et al., 2022). Studies are focused on organic acid production such as lactic acid, acetic acid, citric acid, propionic acid, fumaric acid and succinic acid (Das et al., 2015; González-García et al., 2018; Parmar & Rupasinghe, 2013; Piwowarek et al., 2019; Shalini & Gupta, 2010). Production of these organic acids from apple pomace holds the potential to decrease petrochemical use by either producing them from biomass instead of the commercial petrochemical raw material or by being a biobased alternative to some petrochemicals. Apple pomace is studied to create 3D items like fiberboards and biofilms as well as to produce various biopolymers such as

polyhydroxyalkanoates (PHAs) (Gołębiewska et al., 2022). It can also directly be used to extract dietary fibers, xyloglucan, natural antioxidants, and aromatic compounds (Awasthi et al., 2021). For the environmental impact of apple juice, a study pointed out the biggest impacts were caused by glass packaging (Le Féon et al., 2023). Another study revealed that the apple cultivation stage had more effect on the environmental impact, than the juicing process (Cheng et al., 2022).

2.2.1. Succinic Acid

Succinic acid ($(CH_2)_2(CO_2H)_2$) is a four-carbon organic acid, known as butanedioic acid, 1,2-ethanedicarboxylic acid, and amber acid (Nghiem et al., 2017). It is naturally found as a typical metabolite by plants, animals and microorganisms. (Saxena et al., 2017). Succinic acid was first purified by the mineralist and Doctor Georgius Agricola in 1546, by using dry distillation (Saxena et al., 2017). Nobel Prize winner Dr. Robert Koch recognized the health benefits of succinic acid and introduced succinate as an industrial organic compound in the food industry, generating significant interest in its mass production. (Sharma et al., 2020). Since then, it has been used in multiple industries such as pharmaceutical, agriculture and food (Saxena et al., 2017). Annually, the global demand is nearly 40,000 – 45,000 tons (Sharma et al., 2020). In 2021, the estimated market value for bio-based succinic acid was 110.4 million USD, with the overall succinic acid market (including both bio-based and fossil-based succinic acid) reaching 222.9 million USD (Kim et al., 2023).

Succinic acid is commonly used as a surfactant, detergent and foaming agent for cosmetics and personal care products, as well as household chemicals (Sharma et al., 2020). It can be used as a chain extender for polymeric applications and as an ion chelator to prevent corrosion (Vinoth Kumar et al., 2016; Zeikus et al., 1999). For food applications, it is used in flavoring and fragrance, pH modifier, anti-microbial agent (Vinoth Kumar et al., 2016). It can be utilized as a plant growth regulator, fungicide and herbicide for agricultural applications. It is also used in the production of various chemicals. It is used to produce biodegradable polymers for clothing fibers, and biodegradable plastics, and to produce multiple chemicals such as resins, inks, coating materials, photographic chemicals, dyes, plasticizers and lacquers (Saxena et al., 2017; Vinoth Kumar et al., 2016). It is also used in health-related products such as antibiotics, pharmaceuticals, vitamins and amino acids for pharma-industries (Zeikus et al., 1999). Succinic acid as a commodity chemical, has the potential to substitute various benzene-derived products and intermediate petrochemicals,

leading to a decrease in both the pollution from manufacturing and using over 250 chemicals derived from benzene (Zeikus et al., 1999).

Succinic acid, fumaric acid and malic acid are gathered under the 1,4-diacids and NREL identified them as one of the twelve building chemicals that can be biologically or chemically produced from sugars. These building blocks are important since they can be converted into various high-value bio-based chemicals. Succinic acid is a starting material for valuable chemicals such as THF, 1,4-Butanediol, γ-butyrolactone, pyrrolidones, esters, diamines, 4,4-Bionelle, hydroxybutyric acid (Werpy & Petersen, 2004). To understand succinic acid as a platform chemical, it's beneficial to study its derivatives; succinate esters and succinite anhydride. Succinate esters act as a precursor for 1,4-butanediol, tetrahydrofuran, and γ-butyrolactone and succinic anhydride as fumaric acid and maleic acid (Takkellapati et al., 2018). Here, 1,4-butanediol (BDO) has the largest market application since it is an industrial chemical with multiple uses. It serves as a solvent to produce various chemicals and can also be combined with succinic acid to produce biodegradable plastics like polybutylene succinate (PBS) and poly (butylene succinate-co-butylene terephthalate) (Nghiem et al., 2017). Polyethylene succinate (PES), polypropylene succinate (PPS) and polybutylene succinate (PBS) have been studied extensively, and PES and PBS are commercialized industrially (Takkellapati et al., 2018).

Succinic acid is conventionally produced from the oxidation of butanediol or catalytic hydrogenation of maleic acid, which is a petroleum-derived chemical (Takkellapati et al., 2018). The production process of succinic acid through petrochemical route involves the catalytic oxidation of n-butane to produce maleic anhydride. The maleic anhydride is then catalytically hydrogenated, leading to succinic anhydride, which is further hydrated to produce succinic acid (Smidt et al., 2015). The hydrogenation of maleic anhydride occurs under high temperatures between 120 – 180 °C and high pressure between 0.5 – 40 MPa with the aid of metallic catalysts based on Nickel or Palladium (Pinazo et al., 2015). However, the use of metallic catalysts creates toxicity and increases the overall cost of process (Sharma et al., 2020). Furthermore, it is an energy-intensive process leading to GHG emissions and cost increase. Concerns about oil being a limited resource with volatile pricing and negative environmental impacts enhanced the importance of using biotechnological routes for environmentally friendly production. Nonetheless, bio-based synthesis of succinic acid has gained attention, dating back to the 1980s when the industrial potential of succinic acid fermentation was discovered (Zeikus et al., 1999). Utilizing renewable biomass for bio-based synthesis of succinic acid has various benefits when compared to chemical

processes, such as lower production cost and a more environmentally friendly process since it doesn't contribute to CO_2 accumulation (Oh et al., 2008).

Demand for biomass-derived products has increased due to the depletion of fossil-based raw materials and their negative environmental impacts. Likewise, using alternative renewable resources as feedstock has also gained popularity to produce valuable industrial chemicals (Sharma et al., 2020). Lately, organic acid fermentations gained attraction, including succinic acid, with a focus on its connection to the utilization of agricultural carbohydrates like corn, cassava, and sugar cane. (Zeikus et al., 1999). Generally, succinic acid is produced using either pure sugars or sugars obtained from crop residues or lignocellulosic materials (Sharma et al., 2020). There are studies on using various renewable resources for succinic acid production, including industrial waste and by-product streams, such as cheese whey, crude glycerol from biodiesel production, sugar cane molasses, apple pomace and wheat milling by-products, lignocellulosic materials, such as sugarcane bagasse, corn fiber, corn cob, and bio-waste cotton, and microalgae hydrolysate (Ferone et al., 2019; González-García et al., 2018). Several companies produce bio-based succinic acid commercially. Key players include BASF, BioAmber, Mitsubishi, Myriant, DSM, Mitsui & Co, Succinity, Roquette Freres S.A, Purac, and Reverdia (Sharma et al., 2020). Myriant uses sorghum, Reverdia uses sugarcane and corn starch, DSM and Roquette use corn starch for their respective processes (Kim et al., 2023).

Microorganisms require carbon dioxide (CO_2) for succinic acid production, which serves as an intermediate product in the tricarboxylic acid (TCA) cycle and as the final product of anaerobic metabolism. There are three distinct pathways for succinic acid production: the reducing arm of the TCA cycle (fermentative pathway under anaerobic conditions), the oxidative branch (under aerobic conditions), and the glyoxylate pathway (under aerobic conditions). The latter two, oxidative and glyoxylate pathways, are typically less preferred because succinate transforms into different forms in those pathways and the anaerobic route is favored since it lacks the enzymes that convert succinate into alternative compounds (Karanfil, 2021). The fermentative routes depend on the selected host, there are naturally occurring sources or genetically modified industrial microorganisms. Naturally occurring hosts include bacteria such as *Actinobacillus succinogenes*, *Anaerobiospirillum succiniciproducens, Mannheimia succiniciproducens* and *Bacillus fragili*, and fungi belonging to the Fusarium, Aspergillus, and Penicillium genera (Smidt et al., 2015). There are studies on engineered *Escherichia coli* to develop recombinant strains that can produce succinic acid (Bechthold et al., 2008). As of today, *A. succinogenes* and *M. succiniciproducens*, isolated

from rumen, are the best candidates for succinic acid production due to their capacity to produce succinic acid as a primary fermentation product (Song & Lee, 2006). When industrial production is assessed, *E.coli* is used by Myriant, *Basfia succiniproducens* is used by Succinity, *Candida Krusei* is used by BioAmber and Mitsui, and *Saccharomyces cerevisiae* is used by Reverdia (Smidt et al., 2015).

Four main methods are available for bioconversion, separate hydrolysis and fermentation (SHF), simultaneous saccharification and fermentation (SSF), simultaneous saccharification and co-fermentation (SSCF) and consolidated bioprocessing (CBP) (Lu et al., 2021). In SHF, cellulose hydrolysis is the first process in which hydrolysates rich in sugar are obtained by various enzymes and fermentation is the second step. In SSF, both hydrolysis and fermentation occur simultaneously, making it a promising strategy (González-García et al., 2018). SSCF is a process built upon SSF, it enables the simultaneous fermentation of pentose and hexose sugars from lignocellulosic hydrolysates within a single reactor. CBP is a newly researched concept that integrates the production of hydrolysates, lignocellulose hydrolysis, and microbial fermentation into a single step (Lu et al., 2021). SSF and SHF have been researched more commonly for bio-based succinic acid production. SSF stands out as a superior process compared to SHF since its practical process integration, simplified experimentation, cost-effectiveness, and the elimination of the need for separate equipment, while also ensuring more efficient production by avoiding end-product inhibition, allowing immediate utilization of the produced sugar by microorganisms for growth and production (Karanfil, 2021). Some studies utilized SSF to produce succinic acid from various biomass such as corn stover, cotton stalk and apple pomace (González-García et al., 2018; Pateraki et al., 2016). The production process of succinic acid through microbial fermentation involves several stages, starting with pretreatment, fermentation, product recovery, concentration and purification. The downstream purification is essential since succinic acid is separated from impurities, and this step consists of over 60% of the total production cost (Song & Lee, 2006). Pretreatment is essential for each feedstock, to enhance yield and eliminate inhibitory compounds. Various pretreatment methods such as acid thermal hydrolysis, alkaline thermal hydrolysis, deacetylation, hot water washing, and enzymatic hydrolysis are available; however, selecting the most suitable technology is challenging due to differences in effectiveness, selectivity, and costs (Dickson et al., 2021). The aim here is to break down complex structures such as cellulose, hemicellulose, and lignin, into simple fermentable sugars. The hydrolysate rich in sugar is then sent for fermentation.

The fermentation performance is affected by several factors such as carbon sources, nitrogen sources, pH regulation, and fermentation configuration (Putri et al., 2020). Different fermentation configurations were explained in the previous paragraph, emphasizing SSF and SHF. In SHF, hydrolysis and fermentation are done in separate steps. Hydrolytic enzymes are first applied in the hydrolysis stage, then the hydrolyzed sugars are fermented in the fermentation stage. The advantage here is that each stage can be optimized accordingly since both steps have different requirements (Lu et al., 2021). However, the elevated glucose concentration creates a higher risk of contamination. To address this problem, SSF is proposed. Here enzymatic hydrolysis and fermentation are conducted simultaneously in a single stage, resulting in lower sugar concentrations and reduced contamination risk (Putri et al., 2020). The single-step fermentation is advantageous; however, the optimization of each step cannot be achieved as it was in SHF. Several studies have indicated that SSF provides a highly effective, efficient and time-saving process, converting more cellulose to glucose compared to SHF. Also, the higher saccharification rate enhances productivity, leading to reduced reactor volume and lower capital costs. In SSF, cellulose is hydrolyzed into glucose and the generated glucose is converted into succinic acid concurrently, eliminating the need for a separate saccharification step and reducing the enzyme use (Akhtar et al., 2014). Nitrogen source is one of the significant factors that affects the fermentation performance, various types have been employed such as wheat bran, beef extract, soy peptone, soybean powder, yeast extract, and corn steep liquor. pH levels are another key parameter since cell activities heavily rely on pH regulation. While most bacterial fermentation thrive at near-neutral pH levels, acidification occurs during fermentation. $MgCO_3$, $NaHCO_3$, and Na_2CO_3 have been researched as pH regulators where $MgCO_3$ performed better (Putri et al., 2020). Operating conditions also depend heavily on the selected microorganism whether it's anaerobic or not.

Downstream purification is crucial to isolate pure crystals by removing impurities (such as proteins and broth) and by-products formed during fermentation. The fermentation broth can first be treated by standard centrifugation or filtration to eliminate cellular biomass. The removal of metabolic by-products, such as acetate and formate if *A. succinogenes* is used, is important to achieve succinic acid crystals of high purity, reaching levels around 99.5% (Pateraki et al., 2016). Several methods are evaluated for purification such as precipitation, extraction with solvents and/or amines, ion exchange, membrane separation, esterification, electrodialysis and reactive extraction (Kurzrock & Weuster-Botz, 2010; Orjuela et al., 2013). Commercially, BioAmber used diammonium succinate and reactive evaporation, Reverdia used direct separation of succinic acid, Myriant used ammonia precipitation and Succinity used diammonium succinate as neutralizer

followed by recycling (Dickson et al., 2021). A high purity (>99 wt%) is essential for creating a marketable product however existing technologies have their advantages and disadvantages, and no single method has been universally recognized as the optimal technique for separating bio-succinic acid. The challenge is to select the most economically and environmentally viable technology. The downstream method is closely dependent on the selected feedstock, pretreatment method and pH regulator (Dickson et al., 2021). Current solutions are developed to address challenges, and through downstream processes like vacuum distillation, reactive extraction, and crystallization, purity levels as high as 99.5% can be achieved (Akhtar et al., 2014).

Biomass holds great potential as a feedstock for bio-based succinic acid production, offering both an innovative process and an alternative to fossil-based routes. However economic and environmental assessments are needed for successful utilization. The companies mentioned above have encountered problems and some of them unfortunately went bankrupt or stopped their production. Life cycle assessment offers a comprehensive analysis of environmental impacts thus there are several studies for bio-succinic acid production. Smidt et al. (2015) compared the LCA of bio-based succinic acid with fossil-based succinic acid, showing that European corn-based succinic acid has a reduced carbon footprint (0.9 kg CO_2 eq/kg) compared to fossil-based succinic acid (1.9 kg CO_2 eq/kg), highlighting its positive role in mitigating climate change impacts and addressing concerns related to the depletion of fossil resources. Morales et al. (2016) did a sustainability assessment with cradle-to-gate LCA approach for different fermentation and downstream methods by using glucose from lignocellulosic biomass or sugar beet by using metabolically engineered *E.coli*, resulting that the most environmentally competitive technology is to use metabolically engineered *E. coli*, resistant to acidic pH during fermentation, combined with reactive extraction as purification method. Shaji et al. (2021) presented a techno-economic analysis and LCA of a novel biorefinery to produce succinic acid from sugarcane bagasse, revealing that the climate change impacts are reduced compared to the fossil-based route, however, the difference was not significant. The primary source of emissions is electricity production, especially the high electricity demand for upstream and downstream processes. Dickson et al. (2021) studied a techno-economic and lifecycle assessment for bio-succinic acid production from different feedstocks, glucose, glycerol, corn stover, and dashi kombu, highlighting that the most environmentally friendly process is the optimal glycerol pathway followed by corn stover, glucose, and dashi kombu and for all the routes utility, pH control reagent, and acidification are the primary contributors to environmental indicators. González-García et al. (2018) analyzed the environmental impact of bio-succinic acid production from apple pomace, indicating the purification process as an environmental hotspot where the use

of organic solvents for extraction and distillation unit for solvent recovery raises the environmental burden.

2.2.2. Xylitol

Xylitol ($C_5H_{12}O_5$) is a naturally occurring- five-carbon sugar alcohol that vegetables and fruits naturally have (Ur-Rehman et al., 2015). It is also produced as a metabolic intermediate in mammalian carbohydrate metabolism (Rafiqul & Sakinah, 2013). It was discovered by the Nobel-winner German scientist Emil Fischer in 1890 when he obtained it as syrup from beech wood (Delgado Arcaño et al., 2020). When purified, it is a water-soluble white crystalline powder with a similar taste and sweetness to sucrose, but it has lower calories (Felipe Hernández-Pérez et al., 2019). Xylitol was utilized widely as a sugar substitute in the serious sugar shortage experienced during World War II (Umai et al., 2022). The interest in commercializing xylitol as a sweetener started subsequently (Delgado Arcaño et al., 2020). "Turku Sugar Studies" showed the dental benefits of xylitol in the 1970s and since then numerous products containing xylitol as a substitute have been introduced worldwide (Umai et al., 2022).

Xylitol has applications in the food, dental and pharmaceutical industries thanks to its unique properties (Ur-Rehman et al., 2015). It is used as a low-calorie artificial sweetener for confectionery products, offering several advantages such as minimal impact on insulin release, making it a preferred sugar alternative for people aiming to lower their glycemic index through low-carbohydrate diets (Peterson, 2013). It is a suitable sweetener for diabetic individuals since its breakdown doesn't rely on insulin metabolism and it also helps to treat hyperglycemia (Rafiqul & Sakinah, 2013; Ur-Rehman et al., 2015). Xylitol is used in chewing gums, toothpaste and mouthwash since it prevents dental cavities (Rafiqul & Sakinah, 2013; Umai et al., 2022). It is used as a sweetener or coating agent in pharmaceutical industries (Rafiqul & Sakinah, 2013). It offers a safe and effective treatment for a common ear infection, acute otitis media, believed to work by inhibiting bacterial growth and preventing bacterial attachment to the respiratory tract's surface (Mathur et al., 2023).

According to the NREL Report, xylitol is listed as one of the twelve building block chemicals that can be obtained from sugar through biological or chemical transformations, which can then be converted into various valuable bio-based chemicals (Werpy & Petersen, 2004). Xylitol is a valuable platform chemical with a biological origin that can be used to produce chemicals that have

a petrochemical origin, such as propylene glycol, ethylene glycol, glycerol, xylaric acid, lactic acid and hydroxy furans (Delgado Arcaño et al., 2020). Thus, xylitol serves as a valuable chemical precursor for multiple industries, which contributes to its widespread commercialization and large-scale production with the global market estimated at 200,000 metric tonnes annually by the US Department of Energy (Mathur et al., 2023).

The current commercial production of xylitol is through a chemical process, involving the catalytic hydrogenation of xylose, extracted from lignocellulosic biomass, into xylitol (Umai et al., 2022). Birch wood is used at the industrial level for this process, which is based on the reduction of d-xyloses to xylitol using dilute acids and metal catalysts under high pressure and temperature (Espinoza-Acosta, 2019). This chemical process can be summarized as follows:

-Acid hydrolysis of lignocellulosic biomass to obtain sugar monomers
-Pretreatment of hemicellulosic hydrolysate to purify xylose
-Catalytic hydrogenation of xylose by using Raney-Nickel catalyst under high temperature
-Xylitol purification and crystallization (Delgado Arcaño et al., 2020; Umai et al., 2022)

This is an intensive process with high energy consumption due to hard operation conditions and multiple stages of hydrogenation and purification (Delgado Arcaño et al., 2020). This leads to not only an environmental burden but also an economic burden since the challenging operating conditions led to a significant price increase for xylitol, making it nearly ten times more expensive than sucrose or sorbitol (Espinoza-Acosta, 2019). Biotechnological production methods for xylitol have gained popularity due to the high production cost and environmental disadvantages (Umai et al., 2022). Microbial conversion through fermentation has been studied intensively. In addition to this, xylitol production by using glucose as a substrate and enzymatic conversion of xylose has been considered (Felipe Hernández-Pérez et al., 2019).

The microbial conversion of xylose to xylitol has gained popularity since it offers a straightforward method without relying on toxic catalysts and its production through bacteria, fungi and yeasts has been researched for decades (Ping et al., 2013). The biotechnological approach offers several advantages over the chemical route. These benefits include milder operational temperature and pressure, simplified pretreatment and xylitol purification (Felipe Hernández-Pérez et al., 2019). However, it is also limited by various factors such as temperature and pH of culture, the cost of

nutrients, high water use, and the cost of downstreaming; thus, applying this at the industrial scale can be time-consuming and costly (Rafiqul & Sakinah, 2013).

Lignocellulosic biomass that is rich in hemicellulose can be used as a xylose source, such as wood, agricultural residues and aquatic weeds (Umai et al., 2022). Lignocellulosic biomass is composed of cellulose, lignin, and hemicellulose. Hemicellulose's main component is xylan, a polysaccharide complex that is rich in xylose (Mathur et al., 2023). Since it is a biologically selective process, the production process isn't dependent on purified xylose as a substrate; allowing the use of several hemicellulose-rich biomass to be utilized as a carbon source (Umai et al., 2022). Some studies explored the xylitol production from agricultural wastes such as rice husk, soybean hull, corn cobs, sugarcane bagasse, sorghum bagasse, rapeseed straw, brewer spent grain, wheat straw and cotton straw (Espinoza-Acosta, 2019; Mathur et al., 2023; Sapci, 2012; Vollmer et al., 2023).

The biochemical route involves pretreatment, fermentation, purification and separation steps (Shaji et al., 2022). Lignocellulosic biomass is researched extensively as feedstock; in the first step, it is essential to break down cellulose and hemicellulose into fermentable sugars. The hydrolysis of hemicellulosic fractions of lignocellulosic biomass, such as hardwoods and agricultural residues, aims to optimize the recovery of pentose sugars (xylose or arabinose), minimize the formation of toxic and inhibitory byproducts and to create efficient and cost-effective conversion (Rafiqul & Sakinah, 2013). Various pretreatment methods can be applied using concentrated acid, dilute acid, alkaline, sulfur dioxide, hydrogen peroxide, steam explosion (autohydrolysis), ammonia fiber explosion, wet-oxidation, lime, liquid hot water, CO_2 explosion and organic solvent treatments (Ur-Rehman et al., 2015). The suitable pretreatment method should be selected according to biomass and its composition. Usually, acidic pretreatment is preferred for hydrolysis. Sulfuric acid is commonly preferred but alternatives such as hydrochloric acid, phosphoric acid, trifluoroacetic acid, acetic acid, maleic acid, succinic acid and citric acid are also utilized (Mathur et al., 2023).

After the pretreatment, hydrolysate includes various products such as glucose, xylose, galactose as well as inhibitors including furfural, hydroxymethyl furfural (HMF), acetic acid and phenolic compounds. Furfurals are the main microbial growth inhibitor for xylitol production. To enhance the fermentability of hydrolysates and eliminate microbial growth inhibitors, various detoxification strategies, including physical, chemical, and biological techniques, have been developed (Ur-Rehman et al., 2015). Saturation with lime, neutralization, solvent extraction, use of

ion-exchange resins, charcoal adsorption, evaporation, microbial cell adaptation and treatments with enzymes such as laccase and peroxidase are the techniques used for detoxification. The charcoal adsorption and the ion-exchange resin have been identified as the most effective methods (Rafiqul & Sakinah, 2013). The detoxification process is dependent on the biomass and its composition as well as the selected microorganism and its properties. Another approach to eliminate the adverse effects of inhibitors is to use microorganisms that can tolerate these compounds. There are natural microorganisms that can tolerate the presence of inhibitors, for example, *D. hansenii* can metabolize xylose when acetic acid and furfural are present (Espinoza-Acosta, 2019). Metabolically engineered microbial strains are also developed to tolerate inhibitors. When resistant microorganisms are used, the detoxification step might be no longer needed (Ur-Rehman et al., 2015).

The fermentation step comes after the detoxification of xylose-rich hydrolysate. Simple sugars including xylose, glucose and arabinose, and other carbon sources such as organic acids and oligosaccharides can be used as a carbon source for fermentation. The utilized carbon source differs according to the selected microorganism (Mathur et al., 2023). The biotechnological synthesis of xylitol relies on microorganisms to convert xylose into xylitol, utilizing bacteria, fungi, yeast and genetically engineered strains to produce xylitol from either pure xylose or a hemicellulosic hydrolysate (Rafiqul & Sakinah, 2013). Yeasts are considered the best xylitol producer microorganisms. There are many studies for yeasts that naturally ferment xylose such as *Candida boidinii, Candida guillermondii, Candida tropicalis, Debaryomyces hansenii, Pachysolen tannophilus* and *Pichia guilliermondii* (Ping et al., 2013).

Fermentation of xylitol relies on various factors, including biomass type, hydrolysis conditions, and fermentation parameters such as pH, substrate concentration, inoculum size, and medium composition (Cunha-Pereira et al., 2017). The growth of yeasts is generally favored in an acidic environment, with xylitol production typically occurring at a temperature of 30°C but the optimum temperature for xylitol-producing yeasts varies between 30 and 37°C according to the selected microorganism. To achieve a high xylitol yield it is crucial to optimize the initial xylose concentration, considering the requirements for biomass type and microorganism (Umai et al., 2022). Various nutrients including phosphate ions might be released during hydrolysis, but usually, a nitrogen source is supplied to the fermentation broth. Organic sources like urea, yeast extract, or rice grain are found to enhance fermentation yields and productivity more effectively than inorganic nitrogen sources such as ammonium sulfate (Gerbrandt, 2014).

Effective downstream processing and recovery of xylitol is essential for a successful production process, by choosing the recovery process determined by factors such as product size, fermentation broth concentration, and identification of impurities (Umai et al., 2022). Recovering high-purity xylitol from the fermentation broth has several challenges due to the presence of impurities, nutrients, and chemicals from biomass and microbial cells. Thus, multiple purification steps are needed, including methods like centrifugation for insoluble fractions and activated charcoal for decolorization (Mathur et al., 2023). Ion exchange resins and chromatography can be utilized for xylitol recovery too (Ur-Rehman et al., 2015). Ion exchange resins, activated charcoal, liquid-liquid extraction and membrane separation are used for the clarification of the fermentation broth. This is an essential step before crystallization, which is the most common xylitol purification method (Umai et al., 2022).

With the increasing interest in alternative xylitol production processes, some research focuses on the environmental impacts. In their research, Hafyan et al. (2019) included an LCA to produce xylitol via chemical hydrogenation by using empty fruit bunches as raw material. According to their results, 3.83 kg of CO_2eq is emitted by 1 kilogram of xylitol produced and the chemical hydrogenation step has a major contribution to nearly all impact categories, due to the heat consumption. Dasgupta et al. (2021) researched the energy assessment and life cycle analysis of xylitol from corncob via fermentation, revealing that the whole process emits 8.68 kg CO_2eq which is mainly caused by fossil fuels used in various stages. Marine aquatic and freshwater ecotoxicity cause the highest environmental burden, coming from corn cultivation. Shaji et al. (2022) made a techno-economic and LCA study for xylitol production from sugarcane bagasse via fermentation and according to the LCA results the climate change impact is 1.807 kg CO_2eq per kg of xylitol, where fermentation and filtration stages cause the most of this impact because of the high electricity use. Vollmer et al. (2023) did an LCA for an integrated xylitol biorefinery from wheat straw via bio-based processes. The results show that the global warming impact of the xylitol biorefinery is minimal while the freshwater toxicity, marine ecotoxicity, and human carcinogenic potential are higher, mostly caused by electricity use.

2.3. Decarbonization

Decarbonization is essential to successfully implement the Paris Agreement as keeping the temperature increase below 1.5 °C. To achieve this, scientists support GHG emission reduction of 45% by 2030 and net zero by 2050 (Lau & Tsai, 2023.) The European Union is committed to

reducing GHG emissions by at least 55% by 2030 within the “Fit for 55” package (European Commission, n.d.-b). Furthermore, the EU strives to be climate-neutral with net zero GHG emissions by 2050 (European Commission, n.d.-c.) Türkiye builds its climate change policy in line with the Paris Agreement and EU policies, especially the EU Green Deal. In the Nationally Determined Contribution (NDC), Türkiye aims to reduce GHG emissions by 41% by 2030, taking 2012 as the reference year and aims to achieve net zero by 2053 (UNFCC, n.d.). The key mitigation policy of Türkiye for energy sector focuses on energy efficiency and renewable energy use for 2030 (UNFCC, n.d.). In the National Energy Plan (2022), Türkiye aims to increase the share of renewable energy sources to 54.8% by 2035 and to 69.1% by 2053 which is in line with EU 2050 Reference Scenarios. New strategies are adopted in Türkiye and globally to achieve these goals. The International Energy Agency prepared a roadmap for energy, to show the ways to reach net zero by 2050, where the green transformation of the electricity sector is crucial. In the net zero emission (NZE) scenario, renewables make up nearly 90% of global generation by 2050 (IEA, 2021).

Agriculture, forestry and land use cause the second most GHG emissions following energy sector (Ritchie & Roser, 2024). The EU aims to reduce the environmental and climate footprint of the food system. The European Green Deal values a sustainable food system and the European Commission aims to achieve the target of “at least 25% of the EU’s agricultural land under organic farming and a significant increase in organic aquaculture by 2030” within the Farm to Fork Strategy. An organic action plan was created to achieve this target (European Commission, n.d.-b). Additionally, the Farm to Fork Strategy aims to reduce the use and risks of chemical pesticide use by 50% and at least a 20% reduction in chemical fertilizer use by 2030 (Maçin, 2021). Various policies and plans have been implemented to manage and control GHG emissions from agriculture in Türkiye since 2015. Türkiye’s mitigation policy for the agriculture sector by 2030 includes optimum nitrogen fertilizer use, increasing manure use in biogas plants, reducing nitrogen fertilizer use by using legumes and using agricultural biomass for energy production (UNFCC, n.d.). To achieve net zero in 2050, agricultural emissions should be significantly reduced. Institute for European Environmental Policy analyzed different pathways to reduce emissions, showing that if these options are combined, the emissions could be reduced by 46% by 2050 without major changes in land use (Lóránt A & Allen B, 2019).

Türkiye Green Deal Action Plan is at the heart of Türkiye’s sustainability actions, focusing on the transition to a sustainable and resource-efficient economy. The plan has 9 main points including

circular economy, clean energy, sustainable agriculture and climate change. Within the actions, it is aimed to prepare National Circular Economy Action Plan (Ticaret Bakanlığı, 2021). Türkiye aims to prevent and reduce waste generation in line with circular economy principles, increase the recovery rate of methane from biodegradable waste and transform wastewater treatment facilities into biorefineries until 2030 as a part of mitigation policy. Also, it is aimed to reduce the landfilled waste percentage in line with reaching the target for zero municipal waste landfilling without pretreatment by 2053 (UNFCC, n.d.). Additionally, the "Green Growth Technology Roadmap" is under preparation to promote green production and R&D studies for new technologies, to support green transition in the chemicals sectors along with other sectors (UNFCC, n.d.).

3. MATERIALS AND METHODS

The life cycle assessment (LCA) methodology was based on the ISO 14040 and ISO 14044 standards (ISO 14040, 2006; ISO 14044, 2006). LCA methodology was applied to identify environmental impacts throughout the production of d-limonene and pectin from waste orange peel from orange juice production, and succinic acid and xylitol from waste apple pomace from apple juice production. Within the scope of this study, future scenarios for valorization processes were created according to the electricity mix for current, 2035 and 2050 scenarios. The landfilling scenario, as the current waste management technique, was also compared with the current valorization scenarios. The overall study was divided into subsystems for each product, including the fruit cultivation, fruit juice production and valorization process stages. There were 4 subsystems for valorization techniques: D-Limonene production from orange peel waste, pectin production from orange peel waste, succinic acid production from waste apple pomace and xylitol production from waste apple pomace. Landfilling is the common practice for these wastes; hence it is given as a scenario. 3 future scenarios by using different electricity mixes were also created.

The functional unit was determined as 1 ton (1000 kg) of fruit juice; orange juice for d-limonene and pectin, and apple juice for succinic acid and xylitol. The required data was obtained from GaBi 8.0 software, the Ecoinvent database and related literature. The environmental impacts were evaluated by using GaBi 8.0 software and CML2001 impact assessment methodology was applied in the following impact categories: abiotic depletion (fossil fuel), acidification potential, eutrophication potential and global warming potential.

3.1. Goal and Scope Definition

This LCA study's goal and scope definition is based on the ISO 14044-2006 standard, including the four phases, the goal and scope definition, the inventory analysis, the impact assessment, and the interpretation phase (ISO 14044, 2006). This study aims to evaluate the environmental impact of the valorization of apple and orange juice wastes through the production of d-limonene, pectin, succinic acid and xylitol. In this study, environmental impacts of landfilling, as the current waste management for fruit juice wastes, valorization routes and future scenarios were investigated through life cycle impact assessment (LCIA). 4 environmental impact categories were selected, and toxicity-related impact categories were not studied within the scope of this study.

Selected impact categories are abiotic depletion fossil fuel (ADP fossil), acidification potential (AP), eutrophication potential (EP) and global warming potential (GWP).

3.1.1. Functional Unit

ISO 14044 standards define functional unit as "quantified performance of a product system for use as a reference unit" (ISO 14044, 2006). For this study, the functional unit is defined as 1 ton (1000 kg) of fruit juice.

3.1.2. System Boundaries

System boundaries are determined according to ISO 14044 standard's definition as "set of criteria specifying which unit processes are part of a product system" (ISO 14044, 2006). 4 subsystems were composed for this LCA study. All system boundaries are given in Appendix B. All the valorization routes were considered as Scenario-1. One subsystem is the d-limonene production from orange peel waste, which includes orange cultivation, transportation of orange to the orange juice factory, orange juice production, transportation of orange peel waste, and d-limonene extraction from orange peel waste given in the Figure 3.1. below.

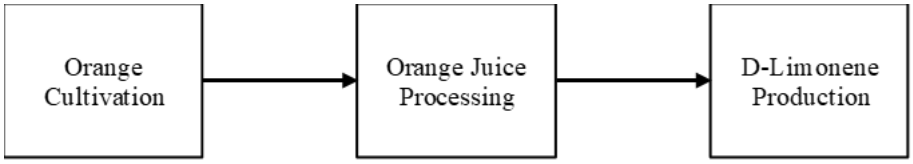

Figure 3.1. Subsystem-1 D-Limonene production from OPW.

The second subsystem is the pectin production from orange peel waste, which includes orange cultivation, transportation of orange to the orange juice factory, orange juice production, transportation of orange peel waste and pectin extraction from orange peel waste by acid hydrolysis given in the Figure 3.2. below.

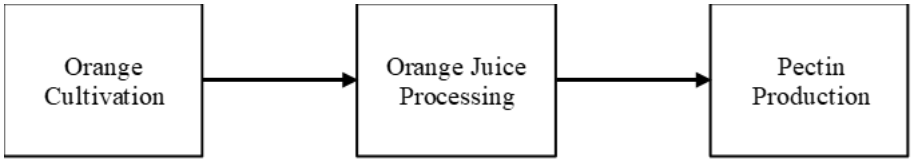

Figure 3.2. Subsystem-2 pectin production from OPW.

The third subsystem is the succinic acid production from waste apple pomace, which includes apple cultivation, transportation of apples to juice factory, concentrated apple juice production, transportation of waste apple pomace and succinic acid production by fermentation; given in the Figure 3.3. below.

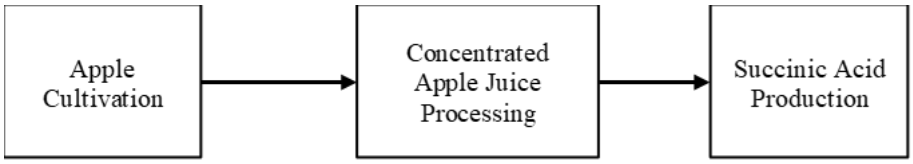

Figure 3.3. Subsystem-3 succinic acid production from WAP.

The fourth subsystem is the xylitol production from waste apple pomace, which includes apple cultivation, transportation of apples to juice factory, concentrated apple juice production, transportation of waste apple pomace and xylitol production from waste apple pomace by fermentation, given in the Figure 3.4. below.

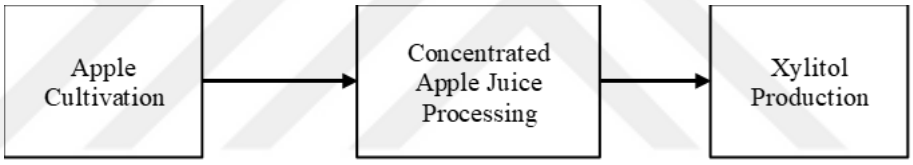

Figure 3.4. Subsystem-4 xylitol production from WAP.

The orange cultivation is adapted from Ortiz-Sanchez et al. (2021), including the germination, nursery, site preparation and planting, vegetative growth, and cultivation stages. The system includes the input data for agrochemicals, irrigation, diesel consumption, and machinery, and the output data for cultivated oranges and emissions (Mariana et al., 2021). The first stage is germination which includes germinator preparation, seed planting, irrigation, and fertilizing. Nursery comes afterward where orange is grafted, including nursery preparation, irrigation, fertilizing, planting and using agrochemicals. In the third stage, the land is prepared for planting. This stage starts with weed control, tree and material removal via machinery, then the seedlings are planted with the addition of fertilizers and agrochemicals. The next step is vegetative growth, in which fertilizers and agrochemicals are used for optimal conditions. In the final step, trees are pruned, fertilizers and agrochemicals are used, and oranges are harvested (Mariana et al., 2021). Harvested oranges are then transferred to the orange juice factory.

The orange juice production process is used according to the standard production. The process includes fruit selection, sizing and grading, washing and juice extraction ("Orange Book", 2023). The process is adapted from Mariana et al. (2021), where the mass and energy balances were based on a data from a real regional company and energy use data from Doublet et al. (2013). Orange juice is produced and waste orange peels are transferred to valorization plants, for d-limonene and pectin extraction.

Substage-1 includes the d-limonene production process, which is adapted from the study of Teigiserova et al. (2022). The process given in the Figure 3.5. includes d-limonene extraction by cold-pressing and solvent-free microwave extraction. Cold press technology is a common method in oil production, employing a mechanical process to apply pressure and puncture the citrus peels to extract the oils (Teigiserova et al., 2022).

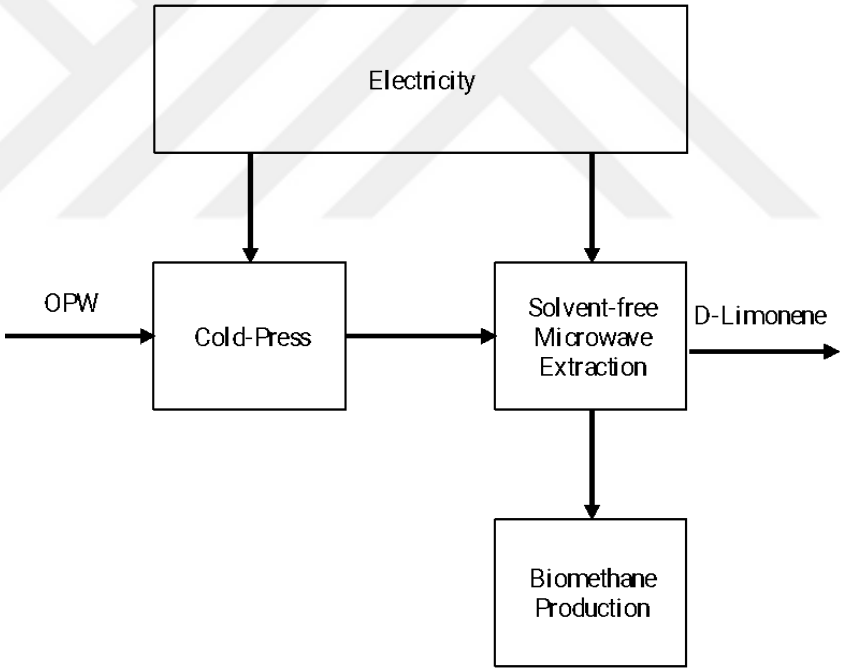

Figure 3.5. D-Limonene production process flowchart.

Substage-2 includes the pectin production given in the Figure 3.6. below. Acid extraction is the most common method, accepted as a conventional method for pectin production (Rodríguez Robledo* & Isabel Castro Vázquez, 2020). The methodology is adapted from studies of Casas-Orozco et al. (2022) and Da Costa et al. (2022). The extraction process includes acid hydrolysis, filtration, precipitation, washing and drying steps.

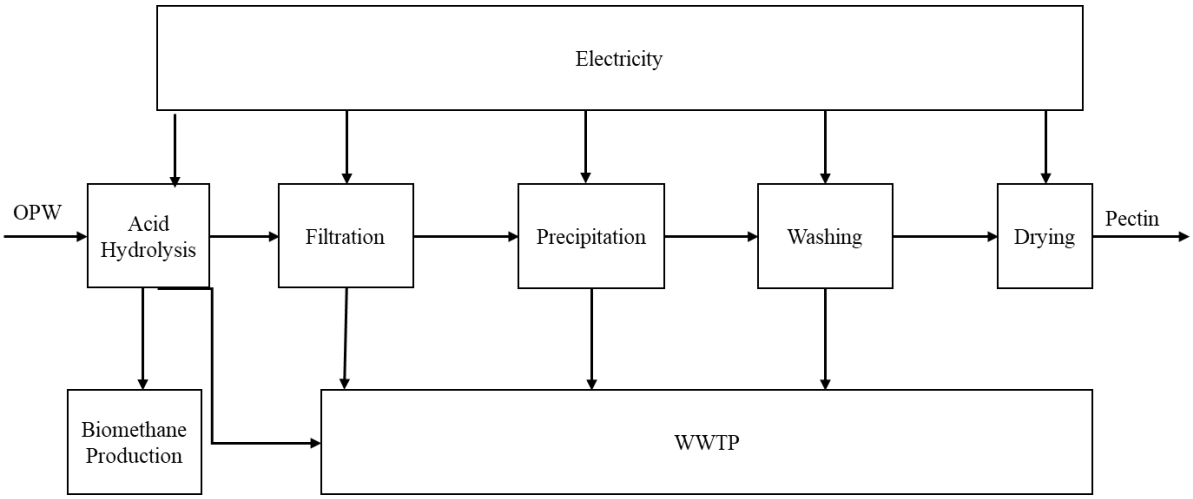

Figure 3.6. Pectin production process flowchart.

Apple cultivation data is adapted from Keyes et al. (2015)'s study. At the farm level, the system includes inputs for land preparation, fuel use of farm equipment, fertilizers, fungicides, insecticides and diesel use for transportation to the orange juice processing factory. The cultivation stages include land preparation, infrastructure, nutrient, pest and disease management, and orchard management.

The apple juice production process is adapted from the study of Cheng et al. (2022). The process includes washing and sorting, crushing, pressing, filtration, enzyme treatment and concentration of the juice. Concentrated apple juice is produced, and the waste apple pomace is sent to valorization plants, for succinic acid and xylitol production.

Substage-3 includes succinic acid production and the process for succinic acid production from waste apple pomace is adapted from González-García et al. (2018) which is based on a full-scale process that is based on the laboratory scale data. Process is divided into 4 main sections: Transportation, Pre-treatment, Fermentation and Purification. Process flow chart is given in Figure 3.7. below.

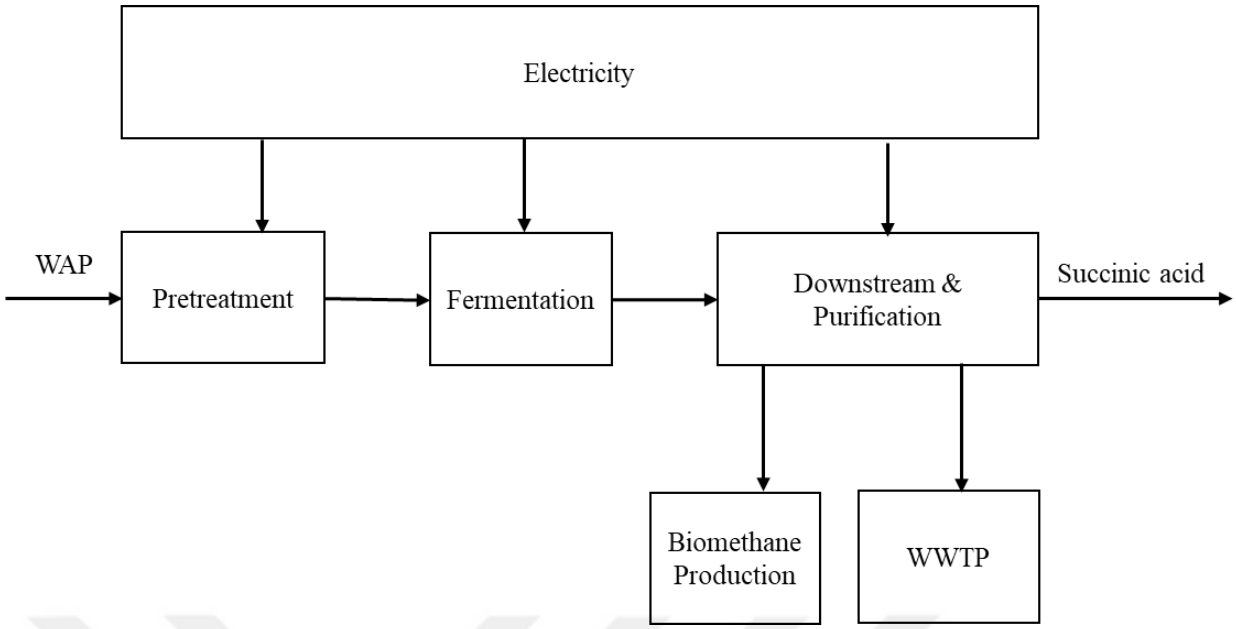

Figure 3.7. Pectin production process flowchart.

Substage-4 includes the xylitol production, which is adapted from Shaji et al. (2022)'s study. The main steps of this process are pretreatment, fermentation and purification given in the Figure 3.8. below.

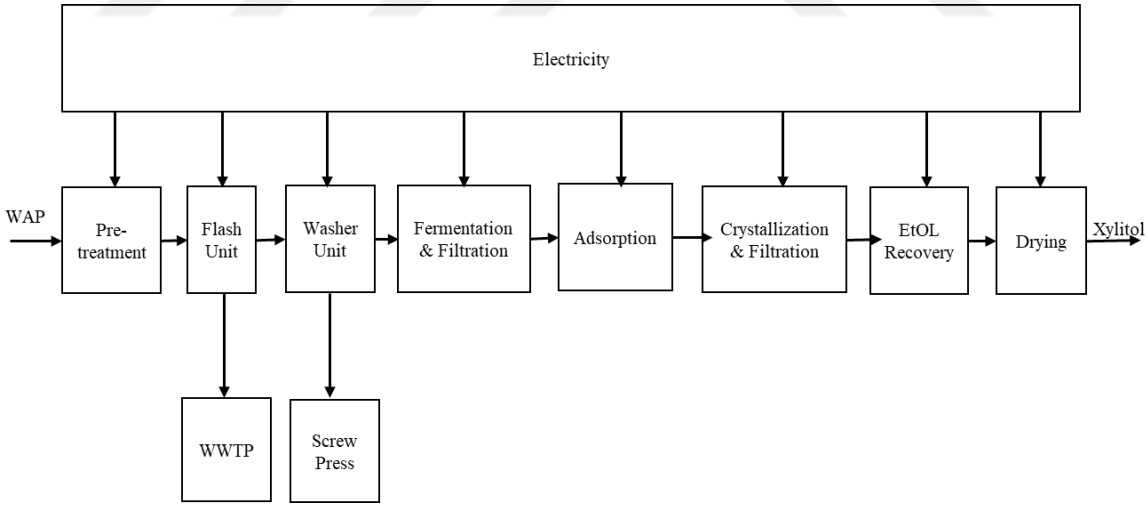

Figure 3.8. Xylitol production process flowchart.

Landfilling is Scenario-2, including orange cultivation, orange juice production and landfilling of the peel waste, and apple cultivation, concentrated apple juice production and landfilling of waste pomace; given in the Figure 3.9. for OPW and in the Figure 3.10. for WAP.

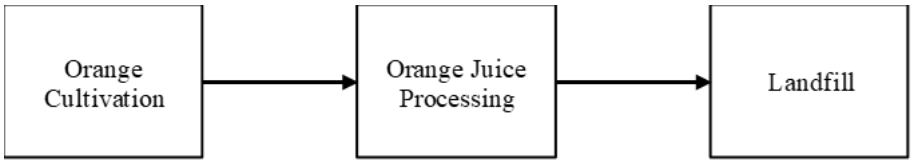

Figure 3.9. Scenario-2 Landfilling of OPW.

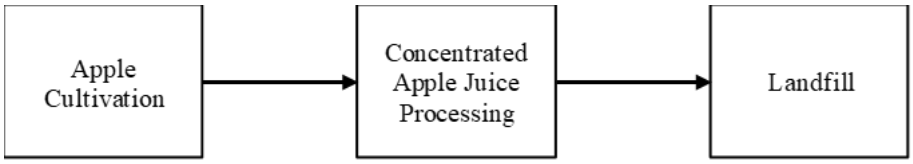

Figure 3.10. Scenario-2 Landfilling of WAP.

3.2. Life Cycle Inventory Analysis and the Key Assumption

Life cycle inventory analysis includes the input and output data for a product through its life cycle. (ISO 14044, 2006). Data that is collected and calculated for this LCA study is given in the Table 3.1. below. The assumptions are given at the end of this section.

Table 3.1. Required data for processes.

Processes	**Required Data**	**Data Sources**
Orange cultivation	Fuel consumption, natural resources, water consumption, agricultural chemicals, emissions	(Mariana et al., 2021)
Apple cultivation	Fuel consumption, electricity consumption, natural resources, water consumption, agricultural chemicals, emissions	(Cheng et al., 2022; Keyes et al., 2015)
Orange juice production	Fuel consumption, water consumption, emissions	(Doublet et al., 2013; Mariana et al., 2021)
Concentrated apple juice production	Fuel consumption, electricity consumption, water consumption, emissions	(Cheng et al., 2022)
Limonene production	Electricity consumption, fuel consumption	(Teigiserova et al., 2022)
Pectin production	Electricity consumption, fuel consumption, water consumption, ethanol consumption	(Da Costa et al., 2022)
Succinic acid production	Electricity consumption, fuel consumption, inoculum preparation, nutrients, enzymes, solvents	(González-García et al., 2018)
Xylitol production	Electricity consumption, fuel consumption	(Shaji et al., 2022)
Biomethane production	-	(Ecoinvent, n.d.)
Landfilling	-	(Mariana et al., 2021)
Electricity	-	(IEA, 2021; T.C. Enerji ve Tabii Kaynaklar Bakanlığı, 2022; T.C. Enerji ve Tabii Kaynaklar Bakanlığı, 2023)

Subsystem-1 and Subsystem-2 start with orange cultivation and orange juice production. 1847 kg orange is used to produce 1 ton (1000 kg) of orange juice, as a functional unit. The agronomic data of orange cultivation is adapted from Ortiz-Sanchez et al. (2021) which covers the Colombian context. The orange productivity is 15.2 tons per hectare in Colombia (DANE, 2016). Whereas in Türkiye the productivity is 30.4 tons per hectare (TÜİK, 2023). The system of Ortiz-Sanchez et al. includes the input and output data for 1 crop hectare equivalent to 18,000 kg oranges cultivated in a grown cycle and the growth cycle is 5.6 years of cultivation, since Türkiye has twice the productivity of Colombia, the cultivation data is adapted accordingly. The used agrochemicals

include mineral fertilizers with nitrogen, phosphorous and potassium, pesticide and inorganic supplements (Mariana et al., 2021).

In the first step of orange juice production, oranges are washed with water and 15% sodium hypochlorite solution. Then oranges are mechanically extracted to obtain orange juice. 1847 kg oranges are used to produce 1000 kg orange juice, the residue is orange peel waste as 918 kg. OPW is transported to d-limonene production and pectin production plants. These plants are assumed to be 40 km to orange juice production site according to orange and orange juice production in Türkiye. According to a pre-feasibility study made for orange oil production in Türkiye, Antalya is selected as the pilot site, thus Antalya is assumed for this study as well. There are 4 orange juice producers in Antalya and 40 km was assumed as the average distance to these producers (Batı Akdeniz Kalkınma Ajansı, 2020).

In Subsystem-1 for d-limonene production orange peel waste is cold-pressed then the final d-limonene is extracted by solvent-free microwave extraction shown in the Figure 3.11. Both steps only use electricity for operation. Left-over orange peel waste is sent to the biomethane production plant. The process scheme on GaBi is given in Appendix A.

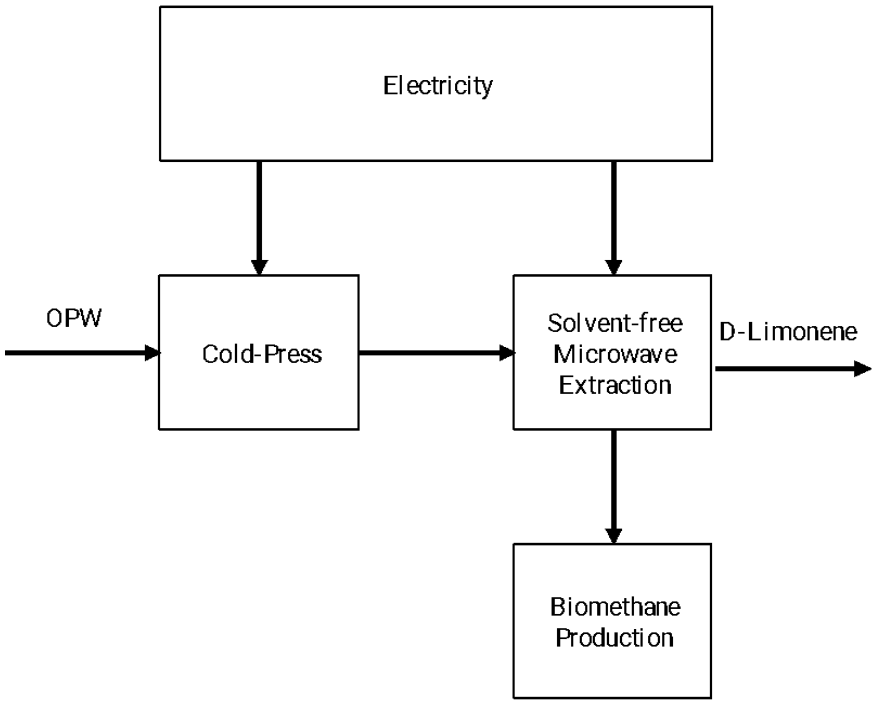

Figure 3.11. D-Limonene production processes flowchart.

In Subsytem-2, the pectin extraction starts with acid hydrolysis of orange peel waste. The literature shows that the highest yield comes from using sulfuric acid, phosphoric acid and hydrochloric acid, and hydrochloric acid is selected for this study. The acid extraction process is carried out at 373.15 K and 2.2 pH for 2 hours (Da Costa et al., 2022; David Durán-Aranguren et al., 2022). Then, the solids are separated from the liquid by filtration using a filter press. The acid effluent goes to wastewater treatment while the solid residue goes to the biomethane production plant. The filtered extract is then precipitated with alcohol, ethanol is selected for this process. The stream with precipitated pectin is washed with water and ethanol to dissolve the pectin. In the final step, the pectin is dried at 40 °C in a dryer to remove water and ethanol, obtaining pure pectin (Da Costa et al., 2022). The leftover orange peel waste is sent to the biomethane production plant. Pectin production process is shown in the Figure 3.12. below. The process scheme on GaBi is given in Appendix B.

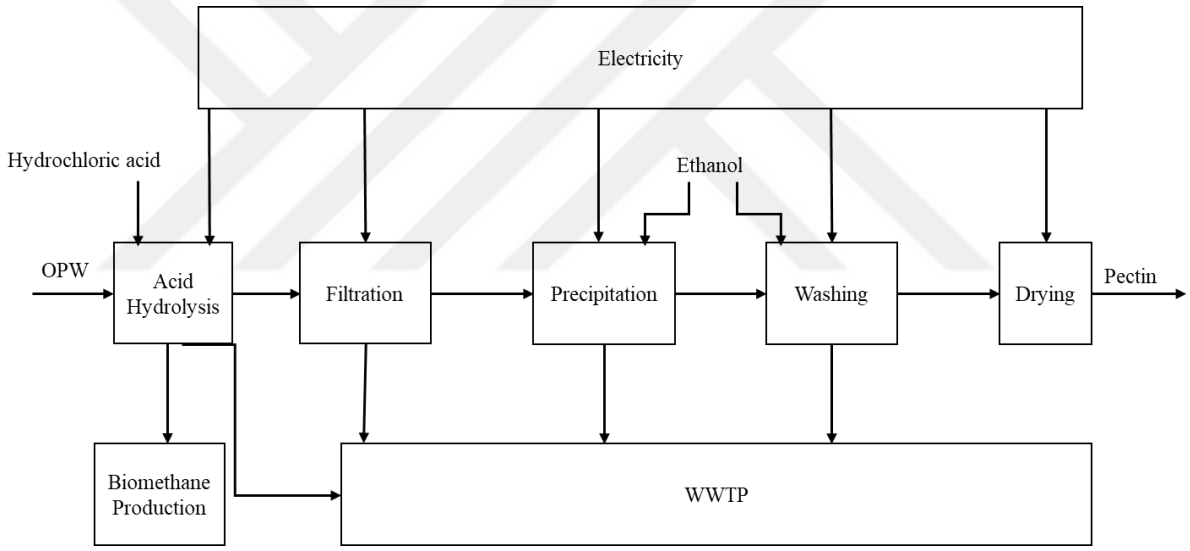

Figure 3.12. Pectin production process detailed flowchart.

Subsystem-3 and Subsystem-4 start with apple cultivation and concentrated apple juice production. 6500 kg of apples are used to produce 1 ton (1000 kg) concentrated apple juice, as the functional unit. The agronomic data of apple cultivation data is adapted from the studies of Cheng et al. (2022) and Keyes et al. (2015). The apple cultivation data of Keyes et al. (2015) was collected from conventional farmers, revealing the apple yield as 23.66 tons/ha, then the data is adapted according to yield in Türkiye which is 26.66 tons/ha (FAOSTAT; Keyes et al., 2015). Minerals, pesticides, mineral fertilizers including nitrogen, phosphorous and potassium, and inorganic supplements are used during cultivation (Keyes et al., 2015).

The apple juice production process and data are adapted from the study of Cheng et al. (2022). The process consists of washing and sorting, crushing, pressing, filtration, enzyme treatment, and concentration steps. After the juice processing, the waste apple pomace is sent to the succinic acid production plant and xylitol production plant. Isparta is the biggest apple producer in Türkiye and there are various apple juice producers as well, thus Isparta is selected as the pilot site. (TEPGE, 2021). These plants are assumed to be 40 km from apple juice production site, just like in orange juice.

For Subsystem-3 in succinic acid production, waste apple pomace is first dried at atmospheric pressure in a tray dryer at 60 °C for 225 minutes. Then the dried apple pomace is stored in silos at 20 °C and atmospheric pressure (González-García et al., 2018). For the fermentation process, *Actinobacillus succinogenes* is selected as the fermentative host. *A. succinogenes* is a wild-type bacterial strain isolated from bovine rumen and it is a natural succinic acid producer. It is one of the most promising strains and one of the most efficient natural succinic acid producers (González-García et al., 2018). Using *A. succinogenes* offers several advantages including its ability to efficiently use various carbon sources, its tolerance against inhibitors, and its capacity to effectively ferment renewable materials. However, it is a fastidious strain that has specific nitrogen, vitamins and pH needs (Pateraki et al., 2016). The nutrient requirements are specified in the process later. SSF process is applied for fermentation since it is a cost-effective single-step process that combines hydrolysis and fermentation under optimal temperature and pH conditions that result in simultaneous conversion into succinic acid; reducing reaction time and costs, and increasing productivity (Akhtar et al., 2014). This section includes the preparation of the inoculum and SSF. In the preparation of the inoculum, *A. succinogenes* cells are grown in a medium that contains glucose and nutrients (yeast extract, corn steep liquor, sodium phosphate monobasic monohydrate ($NaH_2PO_4 \cdot H_2O$), dipotassium phosphate (K_2HPO_4) and magnesium carbonate ($MgCO_3$) (González-García et al., 2018). The cells are recovered through centrifugation after growth, then resuspended in a phosphate buffer solution and inoculated in the fermenter. The SSF medium is prepared by mixing apple pomace and water with a liquid-to-solid ratio of 12 w/w, cellulase, β-glucosidase and nutrients. The inoculum and SSF are performed under the same conditions with a pH of 6.7 (± 0.1), temperature of 37 °C, stirring (150 rpm), and CO_2 flow (0.3 vvm) for anaerobic conditions (González-García et al., 2018). *A. succinogenes* as an anaerobic succinate producer can utilize both glucose and CO_2 as carbon sources and CO_2 is essential for the conversion into succinate during the fermentation of glucose. If integrated into existing biorefineries, the CO_2 released can be captured and reused in this process (González-García et al., 2018).

Downstreaming process is critical to obtain a high-purity product so different methods are researched for the recovery of succinic acid from fermentation broth and purification. After the fermentation step, the output is a solid-liquid mixture so the initial downstream processing involves separating cells through centrifugal separation or microfiltration, typically followed by ultrafiltration to isolate cell debris, proteins, and other polymers from the fermentation supernatant (Kurzrock & Weuster-Botz, 2010). There are different methods evaluated for the purification such as precipitation with calcium hydroxide, electrodialysis, reactive extraction, acidification and esterification in ethanol (Pateraki et al., 2016). The purification process adopted by Garcia et. al. (2018) combines multiple methods including centrifuge, ultrafiltration, reactive extraction, and crystallization. The final product has an industrial grade purity as pure A-grade (≥99.5% wt). The fermenter output is a solid-liquid suspension consisting of succinate salt, formic acid, acetic acid, succinic acid, biomass and residual salts and proteins. It is first sent to centrifuge, solid fraction is recovered as a residue and sent to waste management while the liquid fraction is sent to ultrafiltration membrane unit. The impurities such as biomass, proteins and other salts, are retained in the membranes and the permeate stream is rich in succinate salt, formic acid, acetic acid and succinic acid. To complete the recovery of succinic acid from succinate salt, this stream is mixed with sulfuric acid. Afterwards, reactive extraction is applied by using tri-n-octylamine (TOA) in 1-octanol as dilute in a mixing tank (González-García et al., 2018). Reactive extraction is an effective method for the recovery of carboxylic acids and when it is combined with a recycling process for amines, used as solvents, it has a high potential to be applied in industrial bio-succinic acid production (Kurzrock & Weuster-Botz, 2010). After the reactive distillation, the organic phase includes succinic acid, along with acetic acid and formic acid; so decantation is used to separate organic phase and aqueous phase. Then the organic phase is pumped to vacuum distillation to obtain pure succinic acid. The stream, now rich in succinic acid, is crystallized as the final purification step. The solvents (TOA and 1-octanol) are also recovered and recycled (95%) after the distillation to be reused (González-García et al., 2018). The liquid waste is sent to wastewater treatment plant and the solid waste is sent to biomethane production. The succinic acid production process is given in the Figure 3.13. below. The process scheme on GaBi is given in Appendix C.

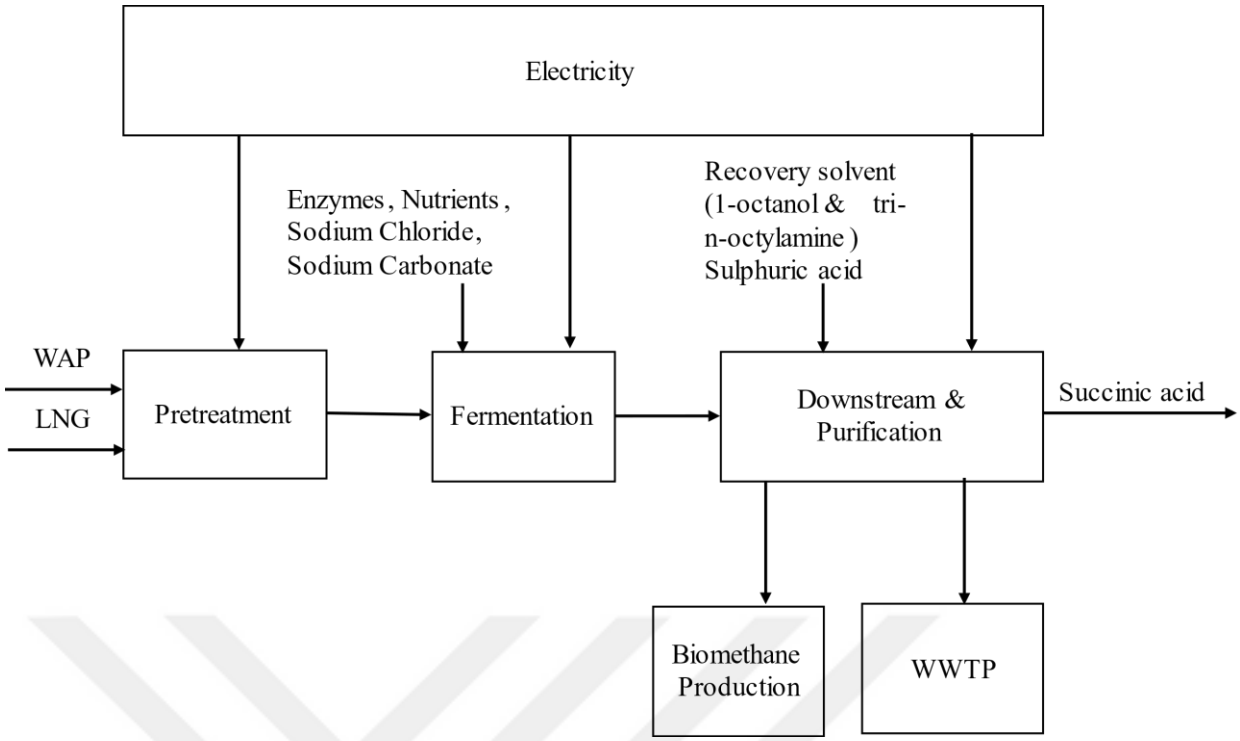

Figure 3.13. Succinic acid production process detailed flowchart.

Subsystem-4 includes xylitol production. The process is adapted for apple pomace from the study of Shaji et al. (2022). Sugarcane bagasse has 26.5% hemicellulose on average while apple pomace has 22.5% on average thus, the data is scaled according to apple pomace (Vaz de Arruda et al., 2017; West, 2021). For pretreatment, dilute acid hydrolysis is selected. Acid hydrolysis is a quick and simple method that is utilized commonly for lignocellulosic biomass. When applied to hardwoods or agricultural residues, it produces a hydrolysate rich in xylose along with small amounts of other sugars. According to the acid concentration, there are two types of acid hydrolysis: Concentrated acid and dilute acid hydrolysis. Concentrated acid hydrolysis uses acids such as H_2SO_4 and HCl, under low operating temperatures (<50 °C) and atmospheric pressure with a high acid concentration of 50-70% v/v. Dilute acid hydrolysis is a widely applied method since it is both effective and cost-effective. It is carried out at high temperatures (160-230 °C) and high pressure (around 10 atm) by using mineral acids at lower percentages. In this process, dilute sulfuric acid and steam at 11 bar pressure is used for pretreatment, at 170 °C for 15 minutes. A flash vessel is utilized after the pretreatment to remove wastewater and toxins that can be detrimental to fermenting microorganisms. The product stream is then washed with water, where the xylose is separated from the solid residue. The stream, now rich in xylose, is sent to fermentation (Shaji et al., 2022).

Yarrowia lipolytica is selected as the fermenting organism. It can't thrive on xylose alone thus glycerol is used as a co-substrate, the cells that grow on glycerol converts xylose into xylitol, with a conversion efficiency of 92-97%. Here in the process, glycerol is used as co-substrate and the fermenter operates at 37 °C and 7.5 pH. The sulfuric acid used at pre-treatment reduces the pH so desired pH is obtained by adding calcium hydroxide ($Ca(OH)_2$). The stream from fermentation includes solid calcium sulfate ($CaSO_4$), formed because of calcium hydroxide, xylitol, unconverted xylose, and water. Liquid and solid phases are separated in the filtration unit, and the liquid stream with xylitol is sent to the adsorption unit. The impurities are removed here by using activated carbon as adsorbent. Then the downstream continues for further separation and purification. It starts with a quadruple-effect evaporative system to concentrate xylitol. The concentrated stream is mixed with ethanol and then fed into a crystallizer to crystallize xylitol. The output stream is sent to the filtration unit to separate xylitol crystals from the ethanol-water mixture from the previous unit. Solid xylitol is sent to the dryer to remove remaining moisture and cool down to obtain pure xylitol (Shaji et al., 2022). The xylitol production process is given in the Figure 3.14. below. The process scheme on GaBi is given in Appendix D.

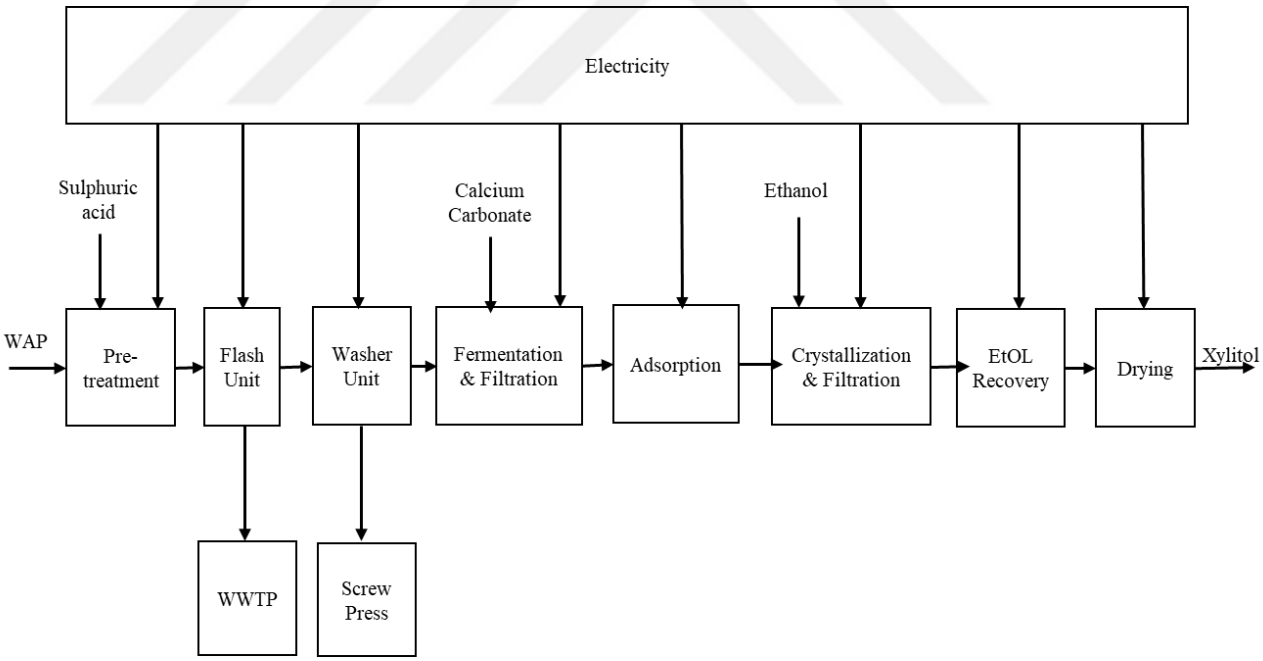

Figure 3.14. Xylitol production process detailed flowchart.

To promote a circular system, leftover biomass after valorization is sent to a biomethane production plant. The unit process of biomethane production from biomass is adapted from the Ecoinvent database (Ecoinvent, n.d.). The electricity use of this plant is not included in the scope.

The landfilling scenario is adapted for orange peel waste and waste apple pomace (Mariana et al., 2021). The electricity mix is created for Türkiye in GaBi, by using electricity production data for 2022 from the Ministry of Energy and Natural Resources. According to this data, 328.3 TWh of electricity was generated in 2022 and the resource share is 34.6% from coal, 22.9% from natural gas, 20.3% from hydropower, 10.6% from wind, 5.1% from solar, 3.4% from geothermal and 3% from other resources, which is assumed as biomass energy (T.C. Enerji ve Tabii Kaynaklar Bakanlığı, 2023).

There are future scenarios designed according to decarbonization goals for Türkiye and the EU. A summary table for all scenarios is given in the Table 3.2 below.

Table 3.2. Summary of Scenarios.

Scenario Name	**Process**	**Agriculture**	**Electricity**
Scenario-1	Valorization	Current (2022)	Current (2022)
Scenario-2	Landfilling	Current	Current
Scenario-3	Valorization	Current	Current + PV
2035 Scenario	Valorization	Farm to Fork Action	Türkiye 2035
2050 Scenario	Valorization	Net Zero – 2050	Net Zero – 2050

Scenario-3 combines the current electricity mix with photovoltaics (PV) for valorization processes since Antalya and Isparta have high potential according to the Solar Energy Potential Map in Türkiye (Enerji İşleri Genel Müdürlüğü, n.d.). It is assumed that half of the electricity need of the valorization plants is met by the grid and the other half is from PV.

The 2035 Scenario includes the combination of 2 variables: Agricultural production for orange and apple are modified according to EU Farm to Fork goals and electricity production was modified according to Türkiye National Energy Plan's 2035 electricity mix. According to the Farm to Fork strategy, chemical pesticide use was reduced by 50%, and fertilizer use was reduced by 20% in 2030, thus the emissions were calculated according to this (European Commission, 2020). 2035 Electricity Mix is created on GaBi according to the following: 20.52% natural gas, 17.70% wind, 17.30% hydro, 16.50% photovoltaic, 13.68% hard coal, 11.10% nuclear, 1.60% geothermal and 1.6% biomass (T.C. Enerji ve Tabii Kaynaklar Bakanlığı, 2022).

The 2050 Scenario includes the 2050 Net Zero agricultural strategy for orange and apple cultivation and electricity production. Multiple actions including improving efficiency and changing production methods, can bring down the emissions by 46% in the agriculture sector by 2050 (Lóránt A & Allen B, 2019). According to the action plan for water efficiency, efficient irrigation systems are promoted in Türkiye (Tarım ve Orman Bakanlığı, n.d.). Drip irrigation is adopted, assuming it decreases the water used for irrigation by 80% (Hillel & Hatfield, 2004). Net Zero Emission (NZE) scenario by IEA (2021) is assumed for the electricity mix and the following mix is created on GaBi: 43.35% photovoltaic, 31.06% wind, 10.90% hydro, 7.97% nuclear, 4.05% biomass, 1.14% geothermal, 0.85% hard coal, 0.68% natural gas and 0.001% heavy fuel oil.

4. RESULTS AND DISCUSSION

4.1. Life Cycle Impact Assessment

In the life cycle impact assessment phase of the LCA, potential environmental impacts of the flows from LCI (ISO 14040, 2006).

4.1.1. Classification

In the classification step, results from the life cycle inventory are designated to the impact categories. In this study, abiotic depletion (ADP fossil), acidification potential (AP) eutrophication potential (EP) and global warming potential (GWP) are selected as impact categories.

4.1.2. Characterization

In the characterization step, each emission type contributes to the relevant impact category and the results are given in the Table 4.1. below.

Table 4.1. Scenario-1 Valorization Impact Categories.

Impact Category	Unit	D-Limonene	Pectin	Succinic Acid	Xylitol
Abiotic Depletion (ADP fossil)	MJ	13053.362	28677.859	68866.608	13636.833
Acidification potential (AP)	kg SO_2-eq.	3.022	6.869	29.915	5.924
Eutrophication potential	kg Phosphate-eq.	0.245	0.577	5.075	1.644
Global Warming Potential	kg CO_2-eq	1408.915	3322.514	8895.722	677.841

Scenario-1 shows the valorization routes. Both d-limonene production (Subystem-1) and pectin production (Subsystem-2) start with orange cultivation and orange juice production. For the

acidification impact category, the main contributors from orange cultivation are emissions of ammonia (NH_3), dinitrogen monoxide (N_2O), nitrogen dioxide (NO_2), nitrogen oxides (NO_x) and sulfur dioxide (SO_2), caused by agrochemical use. For the eutrophication potential category, ammonia, nitrogen oxides and phosphate (PO_4^{3-}) emissions are the contributors, caused by agrochemical use. Energy use is the main cause in all impact categories for orange juice production, in which electricity is the main contributor. Related emissions are given in the Figure 4.1. below for orange cultivation.

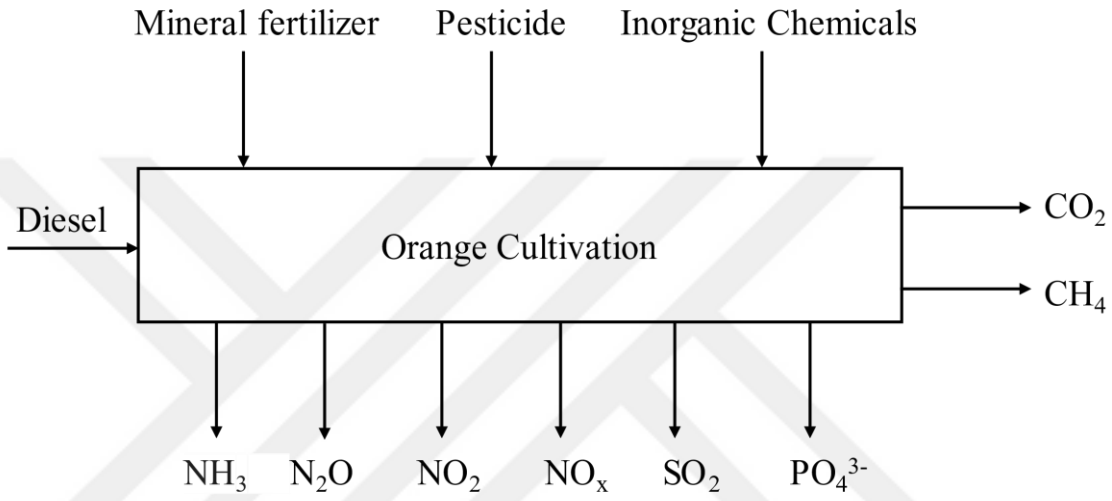

Figure 4.1. Orange cultivation detailed flowchart.

In Subsystem-1, d-limonene extraction comes after the orange juice production. According to the functional unit of 1 ton (1000 kg) of orange juice, the environmental impact of d-limonene production from OPW resulted in 13053.362 MJ, 3.022 kg SO_2-eq, 0.245 kg Phosphate-eq and 1408.915 kg CO_2-eq. This includes the impacts of orange cultivation, orange juice production, d-limonene production, and transportation in between. When the 3 steps are compared, orange juice production has a bigger impact in all categories. Orange juice production contributes to 65% to ADP fossil, 62% to acidification potential, 59% to eutrophication potential and 50% to global warming potential category.

D-limonene extraction process is done in two steps: first the pomace is cold pressed, then sent to solvent-free microwave extraction. While this process eliminates solvent and steam use, it is energy-intensive, thus the emissions are caused by electricity use. Solvent-free microwave extraction uses more electricity than cold-pressing, approximately 23 times more than the cold pressing, hence this unit operation has a bigger impact in all categories, contributing to 96% of

impacts. There are no direct emissions caused by the extraction processes apart from the electricity use. After the extraction, the leftover orange peel waste is sent to the biomethane production plant.

In a study, Pourbafrani et al. (2013) investigated the GHG emissions from producing ethanol, biomethane, limonene and digestate from citrus waste, in two biorefinery scenarios with different sizes. In the small biorefinery scenario d-limonene is the main product, when market value and energy allocation are applied the related GHG emissions are 9115 and 1053 g CO_2eq. kg^{-1} of limonene respectively (Pourbafrani et al., 2013). In this study, according to the functional unit of 1 ton orange juice, 34.85 kg d-limonene can be produced, as a stand-alone product, resulting in 1408.915 kg CO_2-eq. Teigiserova et al. (2022) did LCA for an OPW biorefinery producing d-limonene and citric acid, including the effect of different electricity mixes in Denmark. D-limonene extraction in this study refers to the process in Teigiserova et al. (2022) and it showed that energy-related emissions have the biggest environmental impact.

In the Subsystem-2, the environmental impact of pectin production from OPW resulted in 28677.859 MJ, 6.869kg SO_2-eq, 0.577 kg Phosphate-eq and 3322.514 kg CO_2-eq; including the impacts of the steps from cultivation to valorization. When the 3 steps (orange cultivation, orange juice production and pectin production) are compared, the pectin production step has a bigger impact in all categories, contributing 70% to ADP fossil, 73% to acidification potential, 75% to eutrophication potential and 79% to global warming potential category. The pectin extraction process consists of multiple steps. Acid hydrolysis is the main contributor in all impact categories since 34% of overall electricity consumption comes from this stage. Wastewater exits from all stages except the drying stage and is sent to a municipal wastewater treatment plant and the impact of related emissions is less than 3%. Solid residue from filtration is sent to the biomethane production plant, which contributes to GWP by 25% due to carbon dioxide emissions. The acid hydrolysis step and drying step have the biggest electricity consumption, contributing the most in all categories. The adapted process in this study refers to da Costa et al. (2022) study, in which the hydrolysis unit has the biggest environmental impact for pectin production due to the electricity, hydrochloric acid, and water consumption.

When the two valorization routes are compared, d-limonene production has a lower impact than pectin production. The electricity consumption during pectin extraction is four times the d-limonene extraction. When compared as stand-alone processes, pectin extraction has 4.5 times more impact in all categories than the d-limonene extraction. The study of da Costa et al. (2022) includes

an integrated system for essential oil and pectin production from OPW, showing that pectin production is the main hotspot for environmental effects.

Both succinic acid production (Subystem-3) and xylitol production (Subsystem-4) start with apple cultivation and concentrated apple juice production. Fertilizers, pesticides, and fungicides are the cause of acidification potential and eutrophication potential impact categories. For the acidification potential, the main contributors from apple cultivation are emissions of ammonia (NH_3), dinitrogen monoxide (N_2O), nitrogen monoxide (NO), nitrogen oxides (NO_x) and sulfur dioxide (SO_2); caused by agrochemical use. For the eutrophication potential, the main contributors from apple cultivation are ammonia, nitrate (NO_3^-), nitrogen oxides, and phosphate (PO_4^{3-}) from agrochemical use. For the concentrated apple juice production process dinitrogen monoxide, nitrogen oxides and sulfur dioxide contribute to acidification potential, and nitrogen oxides contribute to eutrophication potential. The concentrated apple juice production process has high electricity use, contributing to ADP fossil and global warming potential impact categories. Related emissions from apple cultivation are given in the Figure 4.2. below.

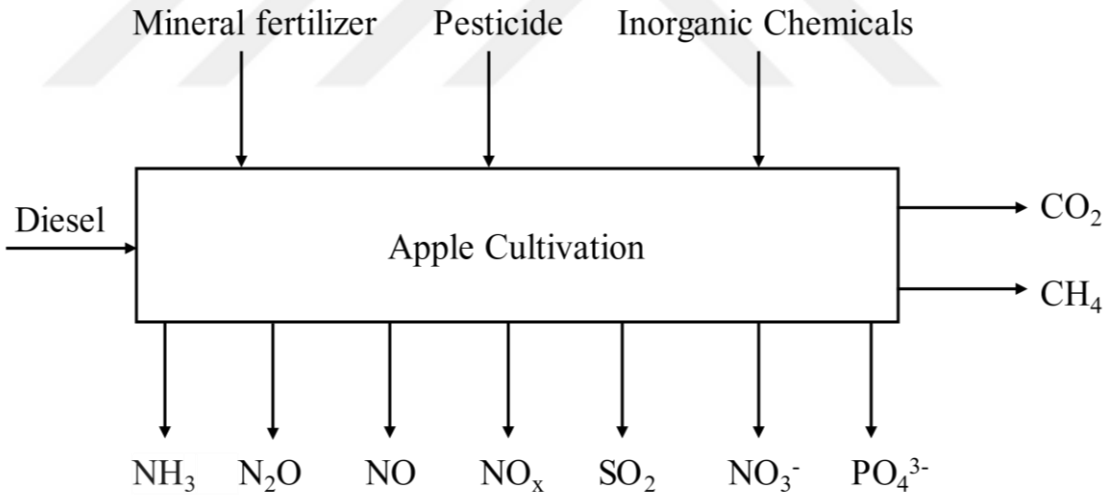

Figure 4.2. Apple cultivation detailed flowchart.

The waste apple pomace (WAP) is treated for succinic acid production in Subsytem-3. According to the functional unit of 1 ton (1000 kg) of concentrated apple juice, the environmental impact of succinic acid production from WAP resulted in 68866.608 MJ, 29.915 kg SO_2-eq, 5.075 kg Phosphate-eq and 8895.722 kg CO_2-eq. When 3 steps are compared; as apple cultivation, concentrated apple juice production and succinic acid production, the succinic acid production step has the biggest impact on all categories.

The succinic acid production process consists of 3 steps: pretreatment, fermentation and downstream and purification. The downstream and purification unit is the most electric-intensive unit consisting of multiple reactors and has high solvent use, hence it has the biggest impact in all categories, contributing to 68% of ADP fossil, 82% of AP, 90% of EP and 77% of GWP. Wastewater and solid waste from downstream units are sent to municipal wastewater treatment plant and biomethane production plant respectively. Only biomethane production has a significant contribution to GWP by 12%, rest of the impacts for both processes are below 2%. According to the LCA results of Morales et al. (2016) covering succinic acid production from various biomass and fermentation processes, energy integration is crucial for lower environmental impacts, compared to conventional petroleum-based production. The study also revealed that reactive extraction as a downstreaming method has the lowest environmental impact, which is the selected method for this study as well. Shaji et al. (2022) revealed a similar result in their study, revealing that the environmental impacts could be improved through improving yield or reducing the energy requirement. Downstream and upstream processes have high electricity consumption and electricity production is the main contributor to emissions. The reference study suggested utilizing bagasse to produce electricity to reduce the grid dependency, which can also lower the overall emissions; as well as utilizing the solid residue in anaerobic digestion (Shaji et al., 2022).

For Subsystem-4 xylitol production from WAP resulted in 13636.833 MJ, 5.924 kg SO_2-eq, 1.644 kg Phosphate-eq and 677.841 kg CO_2-eq. Concentrated apple juice production had the highest impact in the ADP fossil category by 71%, apple cultivation had the highest impact on AP and EP by 77% and %92 respectively. The xylitol production process had the highest impact on the GWP category by contributing 53%. The xylitol production process consists of multiple steps for pretreatment, fermentation, downstream and purification, and for ethanol recovery. Since all types of equipment use electricity, electricity use is the main cause for all impact categories. Flash unit is the most electricity-intensive process, contributing to 65% of EP and 66% of the remaining impact categories. Wastewater from the flash unit is sent to the municipal wastewater treatment plant, however, the impact is way below 1%. Dasgupta et al. (2021)'s LCA study for producing xylitol from corncob, also supports the findings of this study, revealing that chemicals and electricity consumption are among the main drivers of environmental impacts.

When Scenario-1 for succinic acid and xylitol production are compared as stand-alone processes, succinic acid production performs worse environmentally due to its extensive electricity consumption. Succinic acid performs 15 times more in ADP fossil, 26 times more in AP, 45 times more in EP and 21 times more in GWP categories compared to xylitol production.

Scenario-2 is landfilling, as the most common waste management technique for fruit juice waste. Scenario-2 for OPW starts with orange cultivation then orange juice processing and ends with landfilling. The impacts of landfilling (Scenario-2) are compared with valorization routes (Scenario-1) as d-limonene and pectin production and their performance comparison is given in the Figure 4.3. below.

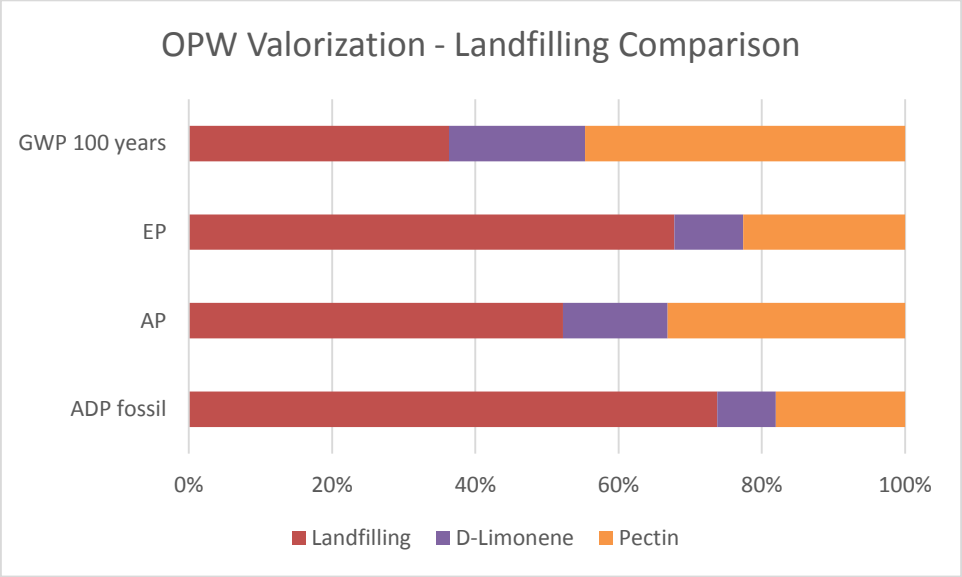

Figure 4.3. Comparison of valorization routes for OPW.

The impacts of landfilling for orange peel waste are as follows: 117423.267 MJ, 10.824 kg SO_2-eq, 1.730 kg Phosphate-eq and 2701.301 kg CO_2-eq. Both valorization routes for d-limonene and pectin are energy-intensive processes however, apart from electricity there aren't any substantial process-related emissions. If d-limonene production is applied instead of landfilling, all impact categories show improved performance: ADP fossil is reduced by 89%, AP is reduced by 72%, EP is reduced by 86% and GWP is reduced by 48%. When landfilling is compared with pectin production, pectin production performed better in all impact categories except GWP. By applying pectin production, ADP fossil is reduced by 76%, AP is reduced by 37%, EP is reduced by 67% but GWP is increased by 23%. Carbon dioxide emission caused by electricity use of pectin production is higher than the emissions from landfilling, hence the impact of GWP increased. Mariana et al. (2021) studied the environmental impact of sanitary landfill disposal and various biorefinery scenarios for OPW, which showed that the environmental impact of landfilling was worse in all categories compared with biorefinery scenarios.

Scenario-2 for WAP starts with apple cultivation, then concentrated apple juice production and ends with landfilling. The impacts of landfilling for waste apple pomace are 226762.409 MJ, 21.479 kg SO_2-eq, 4.509 kg Phosphate-eq and 4027.262 CO_2-eq. The impacts of landfilling (Scenario-2) are compared with valorization routes (Scenario-1) as succinic acid and xylitol production and the results of the comparison are given in the Figure 4.4 below.

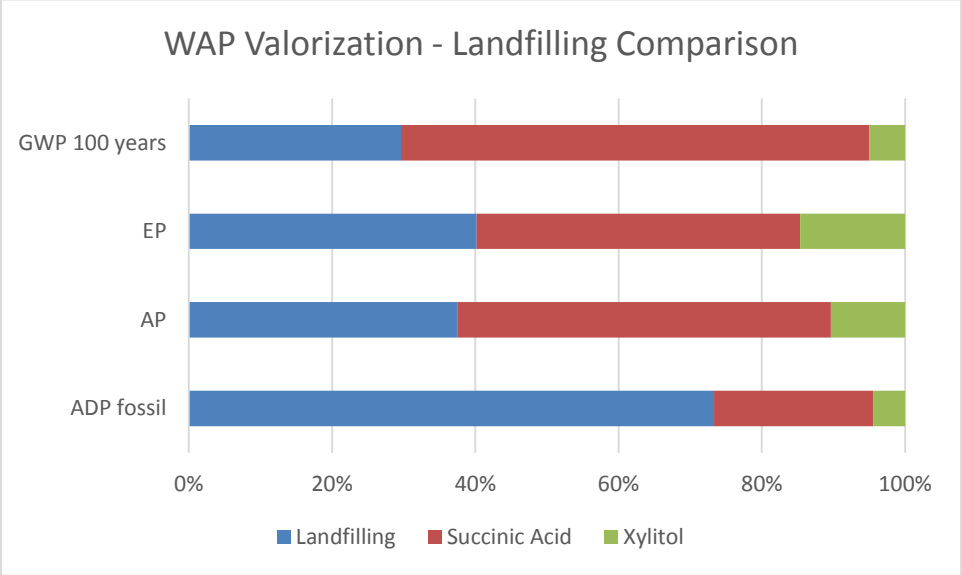

Figure 4.4. Comparison of valorization routes for WAP.

When compared with landfilling, succinic acid performs better only in ADP fossil category and xylitol performs better in all categories. If succinic acid production is applied instead of landfilling, succinic acid production shows 70% reduction in ADP fossil but shows 39% increase in AP, 13% increase in EP and 121% increase in GWP. Due to the amount of renewable energy in the electricity mix, succinic acid production shows better performance in ADP fossil. However, the emissions caused by the extensive electricity consumption surpass the landfilling emissions, creating more impact in the rest of the categories. If xylitol production is applied instead of landfilling, 94% reduction in ADP fossil, 72% reduction in AP, 64% reduction in EP and 83% reduction in GWP are seen. Gassara et al. (2011) analyzed the GHG emissions from apple pomace management techniques, resulting that producing enzyme via fermentation was the most effective method for lower GHG emissions whereas landfilling performed the worst.

Table 4.2. Summary for Future Scenarios.

Future Scenarios	**Process**	**Agriculture**	**Electricity**
Scenario-1 (base case)	Valorization	Current	Current
Scenario-3	Valorization	Current	Current + PV
2035 Scenario	Valorization	Farm to Fork Action	Türkiye 2035 goal
2050 Scenario	Valorization	Net Zero – 2050	Net Zero – 2050

In Scenario-3 (S3) combination of grid electricity and PV is used. All valorization routes performed better in all impact categories. Electricity by PV has lower emissions than the electricity grid mix, so the environmental impacts are lowered. The impact change between Current (2022) and Scenario-3 is given in the Figure 4.5. below.

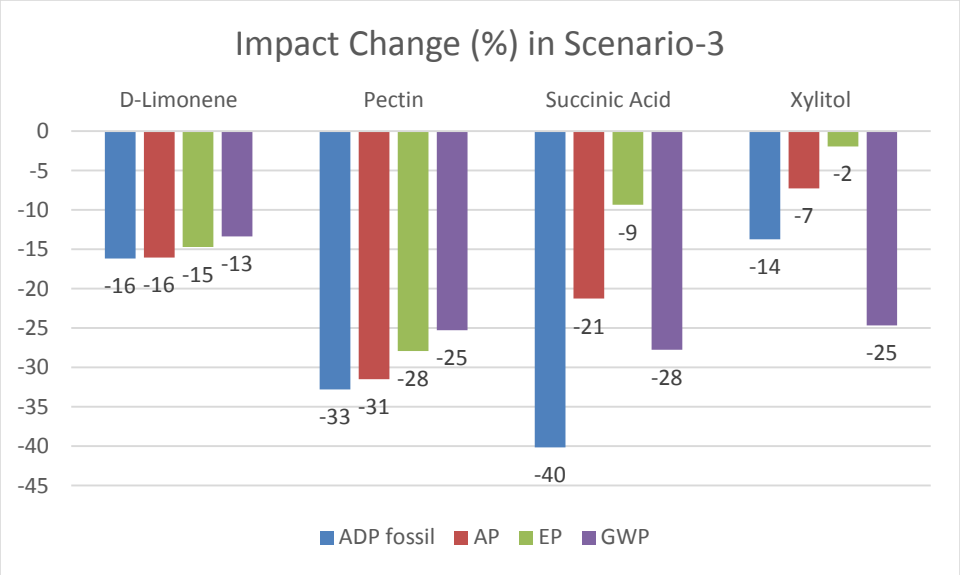

Figure 4.5. Impact change (%) between Current Scenario and Scenario-3.

When the impact changes are compared, d-limonene shows less decrease as the orange juice production is the biggest contributor, but here the renewable electricity only impacts the d-limonene extraction process. In contrast, energy-intensive pectin extraction process is the biggest contributor for this valorization route, thus the impacts improve with the addition of PV. Succinic acid is the most energy-intensive process hence the addition of PV creates the biggest impact in ADP fossil and GWP. EP impact decreases less in succinic acid and xylitol because downstream emissions are the biggest contributor in succinic acid and apple cultivation is the biggest contributor in xylitol, thus the change in electricity only contributes to 9% and %2 reduction respectively.

2035 Scenario included cultivation according to Farm to Fork strategy and targeted electricity mix for Türkiye. For orange and apple cultivation, AP, EP and GWP categories' impacts reduce by 37%. All valorization routes showed reduction in all impact categories in 2035 Scenarios, due to reduced emissions by the increased renewable energy percentage and lower pesticide and fertilizer use. Results are given in the Figure 4.6. below.

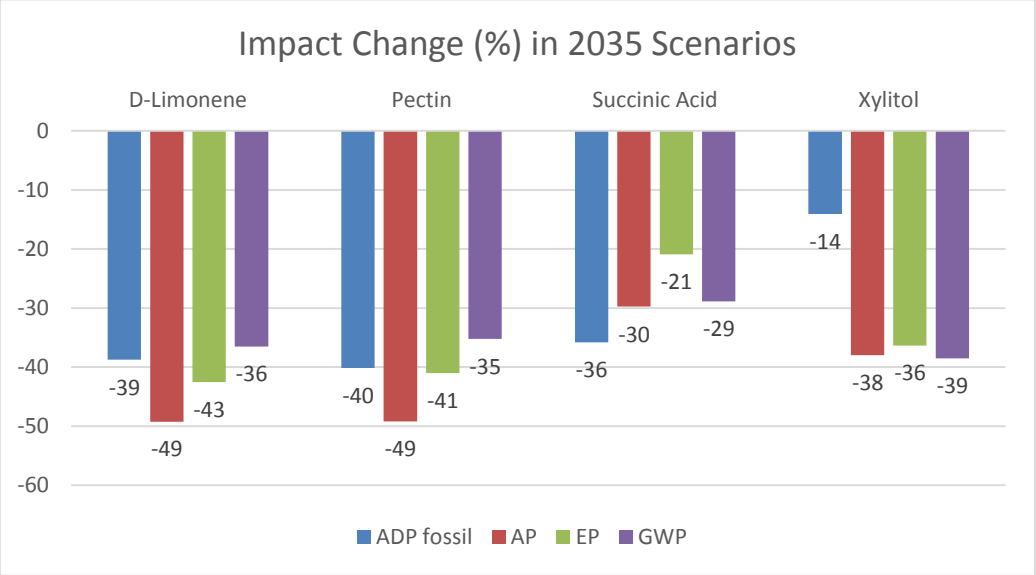

Figure 4.6. Impact change (%) between Current Scenarios and 2035 Scenarios.

For OPW, the changes are similar for d-limonene and pectin, since electricity consumption is the main emission in both orange juice production and extraction processes. For WAP, succinic acid production process is the main contributor for that valorization route and for xylitol concentrated apple juice production is the main contributor for the impact categories. During the apple juice production, coal is used and since this step has the biggest impact, xylitol shows less decrease in ADP fossil. AP and EP categories show more decrease compared to Scenario-3 thanks to lowered agricultural emissions.

2050 Scenario included NZE strategy for agriculture and electricity. Net zero strategy was based on 46% reduction for agricultural emissions so for orange and apple cultivation all impact categories were reduced by 46%. All valorization routes showed reduction in all impact categories when compared to the current scenario. Figure 4.7 below shows the change between current valorization scenarios and 2050 Scenarios.

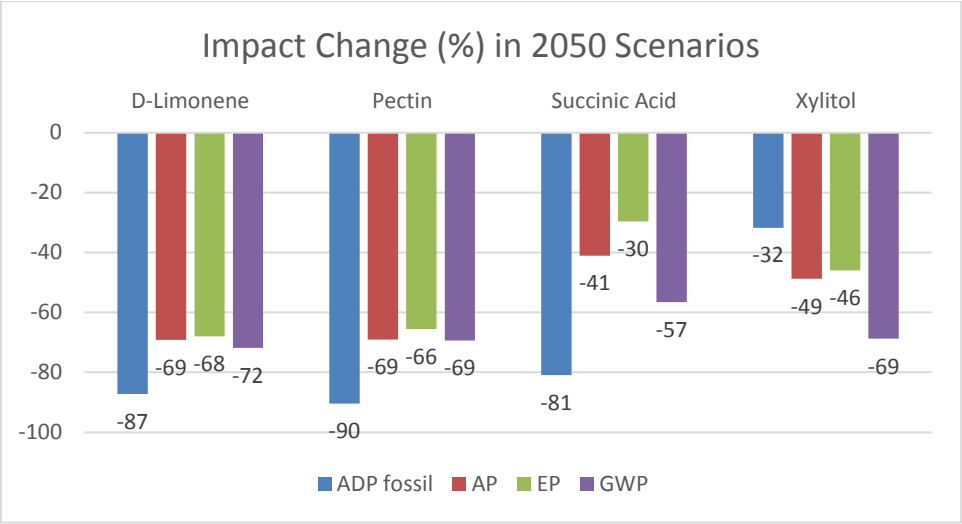

Figure 4.7. Impact change (%) between Current Scenarios and 2050 Scenarios.

D-limonene and pectin show similar results for OPW, showing greater reduction in all categories. Succinic acid and xylitol show less reduction when compared because they have process-related emissions too as opposed to d-limonene and pectin only having major emissions from electricity. Furthermore, the amount of apples used for juicing are 3.5 times more than the oranges to produce the same amount of fruit juice thus agricultural emissions have more impact for WAP valorization routes. The coal use in CAJ production also adversely affects the results. At the EU level, the transition to renewable electricity from fossil had improved in climate change, freshwater eutrophication, particulate matter formation and terrestrial acidification impact categories thus the targeted renewable electricity increase in Türkiye would result in an improved impact (European Environment Agency, 2023).

5. CONCLUSION AND RECOMMENDATIONS

The aim of this study was to identify the potential environmental effects of selected valorization routes for orange and apple juice residues. Orange peel waste was utilized to produce d-limonene and pectin, and waste apple pomace was utilized to produce succinic acid and xylitol. The environmental impacts were identified by using LCA methodology in GaBi 8.0 software by applying CML2001- August 2016. Life cycle impact assessment was applied in four impact categories: abiotic depletion (ADP fossil), acidification potential (AP) eutrophication potential (EP) and global warming potential (GWP). There were 4 valorization routes, landfilling, and three future scenarios. The subsystems were: Subsystem-1 for d-limonene production by cold pressing and solvent-free microwave extraction, Subsystem-2 for pectin production by acid hydrolysis, Subsystem-3 for succinic acid production by fermentation and Subsystem-4 for xylitol production by fermentation. Scenario-1 was 4 valorization routes for the current scenario, Scenario-2 was landfilling for OPW and WAP, Scenario-3 was current electricity mix combined with PV, Scenario 2035 was for future electricity mix and agricultural production and Scenario 2050 was for electricity mix and agricultural production for the NZE scenario. All subsystems and scenarios start with fruit cultivation and fruit juice processing.

For the valorization routes of 4 subsystems, the results were calculated in GaBi. According to the functional unit of 1 ton (1000 kg) of orange juice, d-limonene production and pectin production from orange peel waste were analyzed. 34.85 kg d-limonene and 83.79 kg pectin can be produced from 918 kg of OPW as the residue of 1000 kg of orange juice. In Subsystem-1, d-limonene extraction comes after the orange juice production. According to the functional unit of 1 ton (1000 kg) of orange juice, the environmental impact of d-limonene production from OPW resulted in 13053.362 MJ, 3.022 kg SO_2-eq, 0.245 kg Phosphate-eq and 1408.915 kg CO_2-eq. This includes the impacts of orange cultivation, orange juice production, d-limonene production, and transportation in-between. When the 3 steps are compared, orange juice production has a bigger impact in all categories. Orange juice production contributes to 65% to ADP fossil, 62% to acidification potential, 59% to eutrophication potential and 50% to global warming potential category.

D-limonene extraction process is done in two steps: first the pomace is cold pressed, then sent to solvent-free microwave extraction. While this process eliminates solvent and steam use, it is energy-intensive, thus the emissions are caused by electricity use. Solvent-free microwave

extraction uses more electricity than cold-pressing, approximately 23 times more than the cold pressing, hence this unit operation has bigger impact in all categories, contributing to 96% of impact. There are no direct emissions caused by the extraction processes apart from the electricity use. After the extraction, the leftover orange peel waste is sent to biomethane production plant.

In Subsystem-2, pectin production was the main contributor in all categories. When pectin extraction step was considered, acid hydrolysis step was the main contributor to all impact categories due to the highest electricity consumption and chemical use. Among the two valorization routes, d-limonene performed better since pectin extraction had 4.5 times more impact in all categories.

D-limonene and pectin production are the valorization options considered for OPW in this study. When they are compared with landfilling, as the current waste management technique, d-limonene production performs better in all categories and pectin production performs better in all categories except GWP. If d-limonene production is applied instead of landfilling, all impacts decrease; ADP fossil by 89%, AP by 72%, EP by 86% and GWP by 48%. If pectin production is selected instead of landfilling, ADP fossil is reduced by 76%, AP is reduced by 37%, EP is reduced by 67% but GWP is increased by 23%.

According to the functional unit of 1 ton (1000 kg) of concentrated apple juice, succinic acid and xylitol production from waste apple pomace were analyzed. 898 kg succinic acid and 151 kg xylitol can be produced from 1625 kg of WAP as the residue of 1000 kg CAJ. In Subsystem-3, the succinic acid production step was the biggest contributor to all impact categories. In the succinic acid production process, the downstream and purification step had the highest impact on all categories due to this step having the overall highest electricity consumption by 68% and solvent use. This step contributes to 68% of ADP fossil, 82% of AP, 90% of EP and 77% of GWP categories.

In Subsystem-4 concentrated apple juice production had the highest impact in ADP fossil category by 71%, the xylitol production process had the highest impact on GWP by 53% and apple cultivation had the highest impact on AP and EP by 77% and 92% respectively. The xylitol production process includes multiple steps and electricity use is the main emission cause. Flash unit is the most electricity-intensive process, contributing to more than 65% impact in all categories.

Succinic acid and xylitol production are the selected valorization routes for WAP. If succinic acid production is preferred instead of landfilling, ADP fossil is reduced by 70%, AP is increased by 39%, EP is increased by 13% and GWP is increased by 121%. If xylitol production is applied instead of landfilling, ADP fossil is reduced by 94%, AP is reduced by 64%, EP is reduced by 64% and GWP is reduced by 83%.

Future scenarios consider climate goals for electricity mix and agricultural production. In Scenario-3, electricity use of the valorization process is supplied by the grid and PV by half. The 2035 Scenario includes the targeted electricity mix of Türkiye by 2035 and the implementation of Farm to Fork strategy. 2050 Scenario is based on NZE for electricity and agriculture globally. Since all these scenarios are based on emission reduction, the impact categories greatly improved with these measures. In Scenario-3, using PV reduced the electricity-caused impacts only in valorization processes. As 2035 and 2050 scenarios include a variety of renewables in electricity mix with lowered fossil fuels as well as reduced agricultural emissions; the environmental impact of the valorization routes improved.

The aim of this study was to analyze the potential environmental impact of valorization routes by using LCA methodology. Except for pectin extraction, all routes were novel technologies that need further research and improvement. Through further studies in scale-up strategies, integrated systems such as biorefinery approach and utilizing residues for energy generation integrated into the process to decrease electricity grid use would improve the potential impacts. Türkiye’s target to increase renewable energy share in electricity production is a critical opportunity for emission reduction, thus the valorization routes would perform better with decarbonized electricity production.

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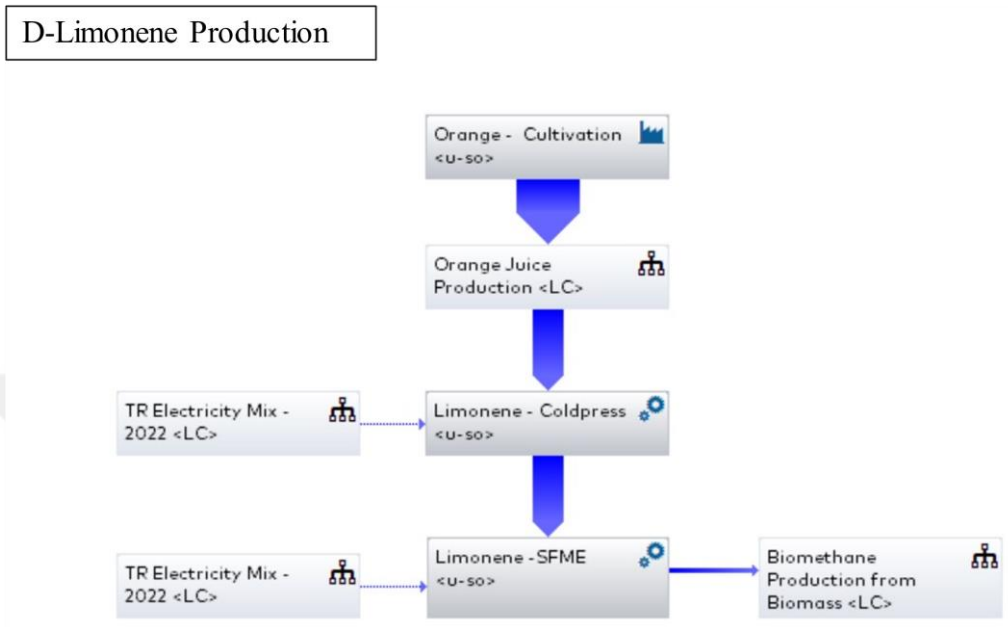
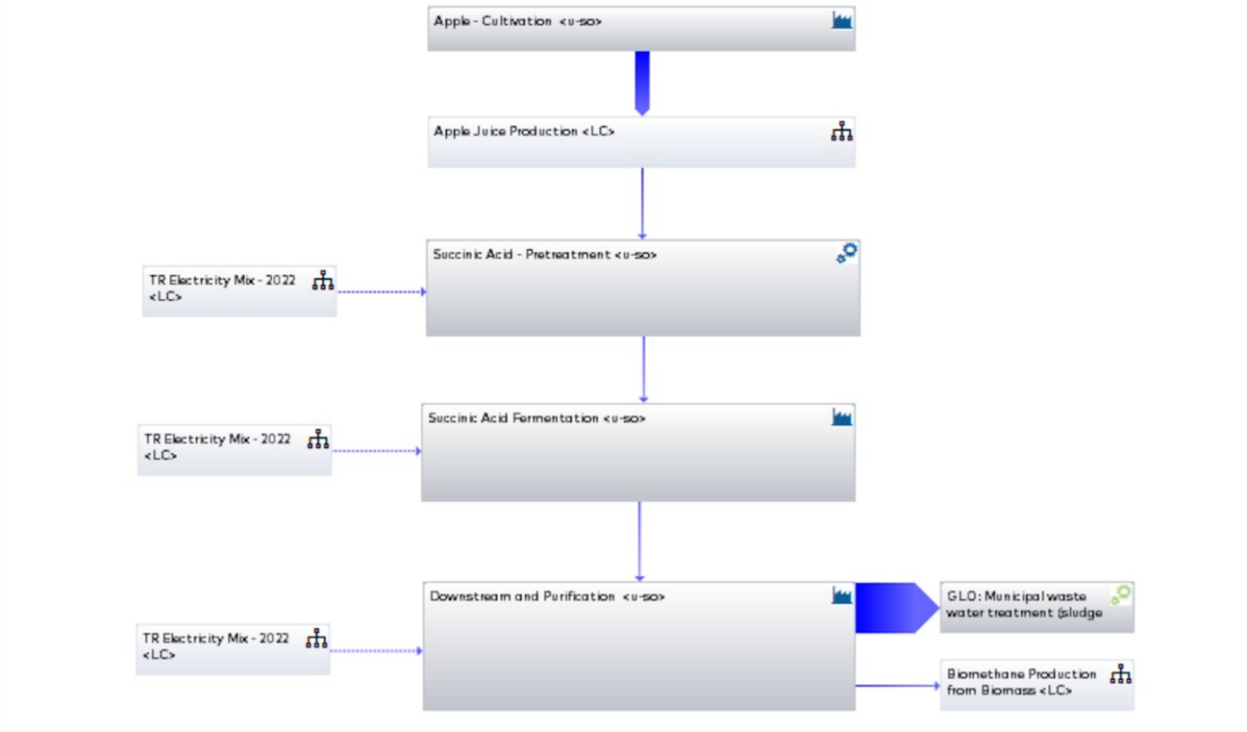
APPENDIX A: PRODUCTION PLANS ON GABI



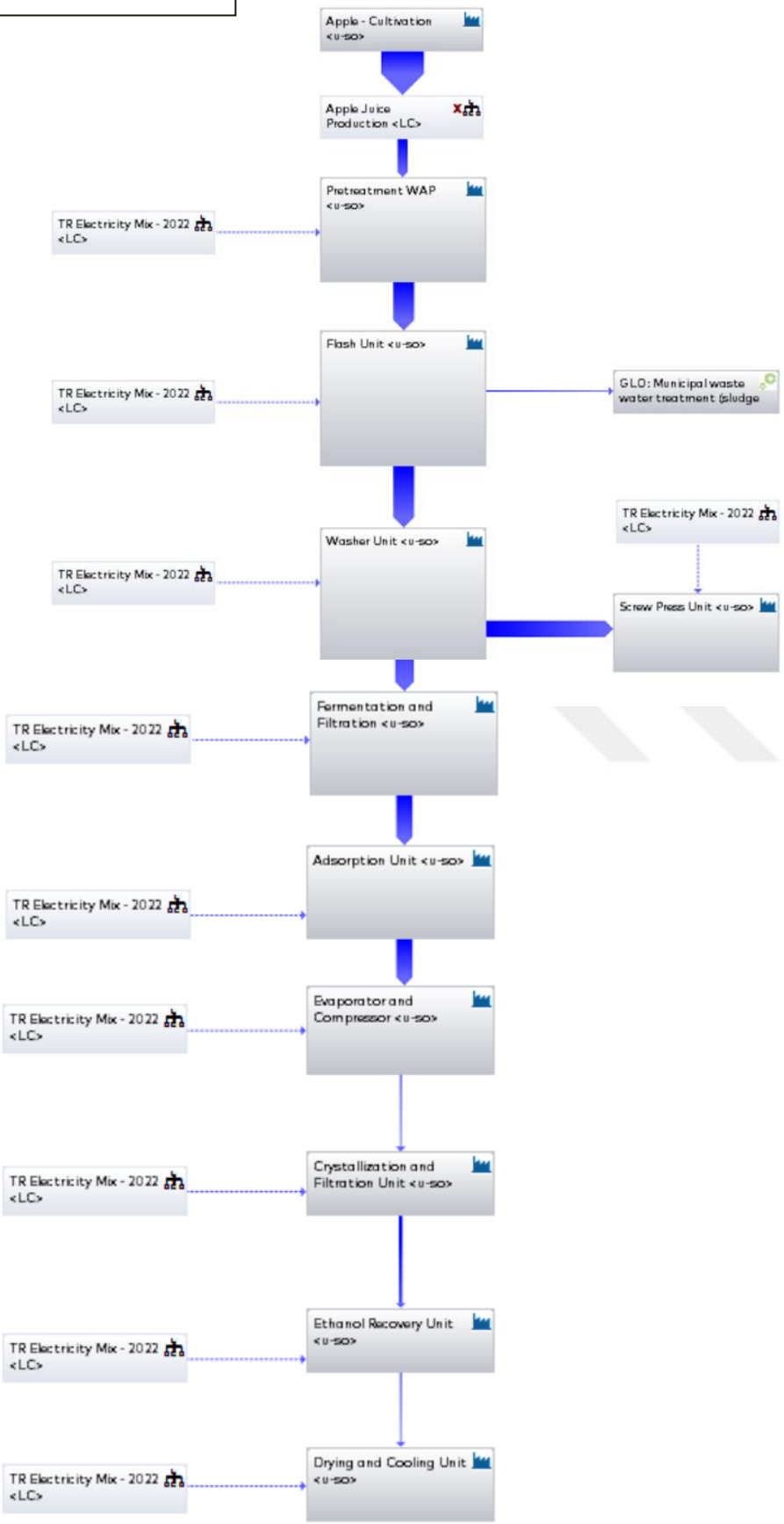
Pectin Production



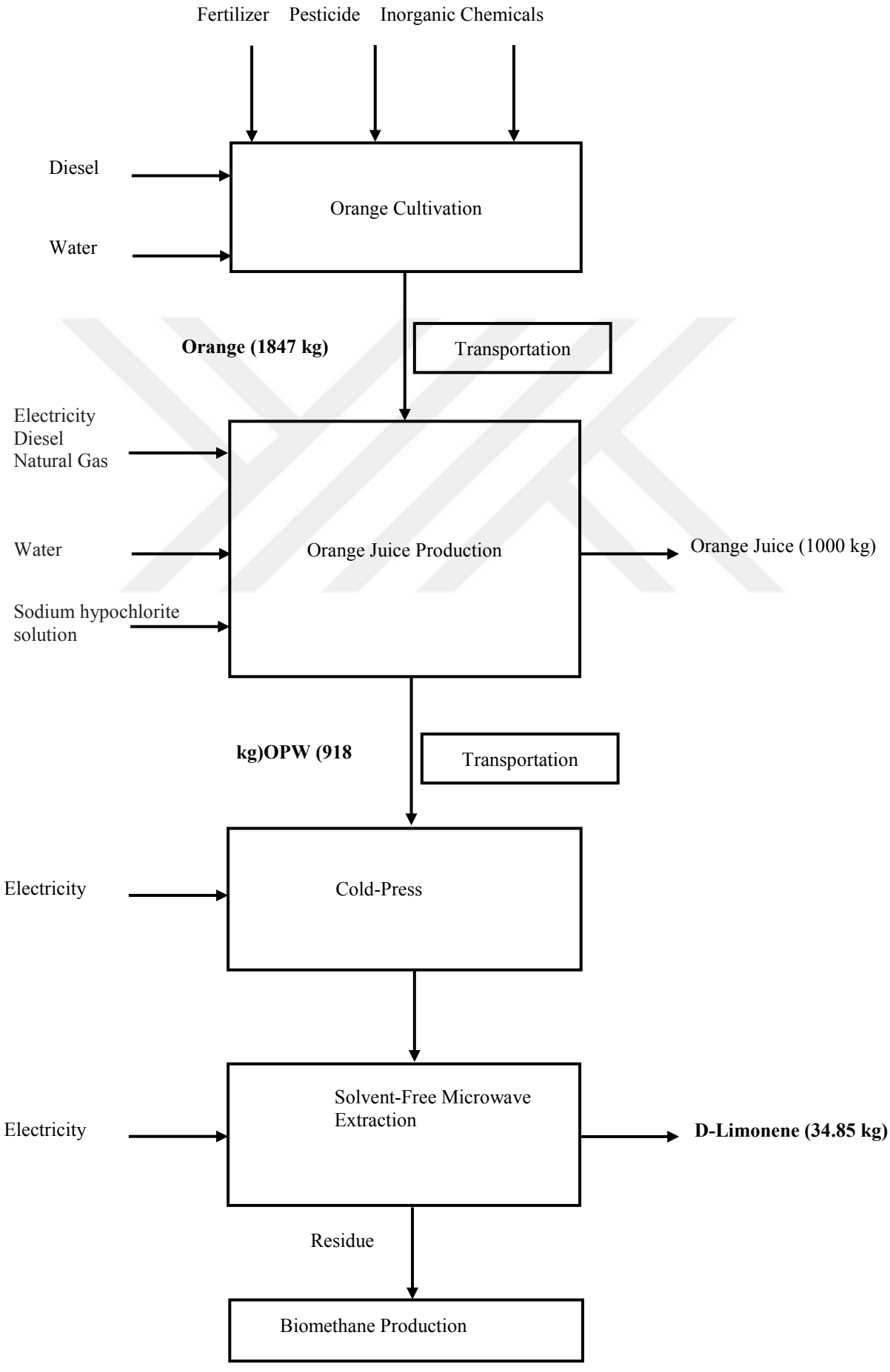
Succinic Acid Production

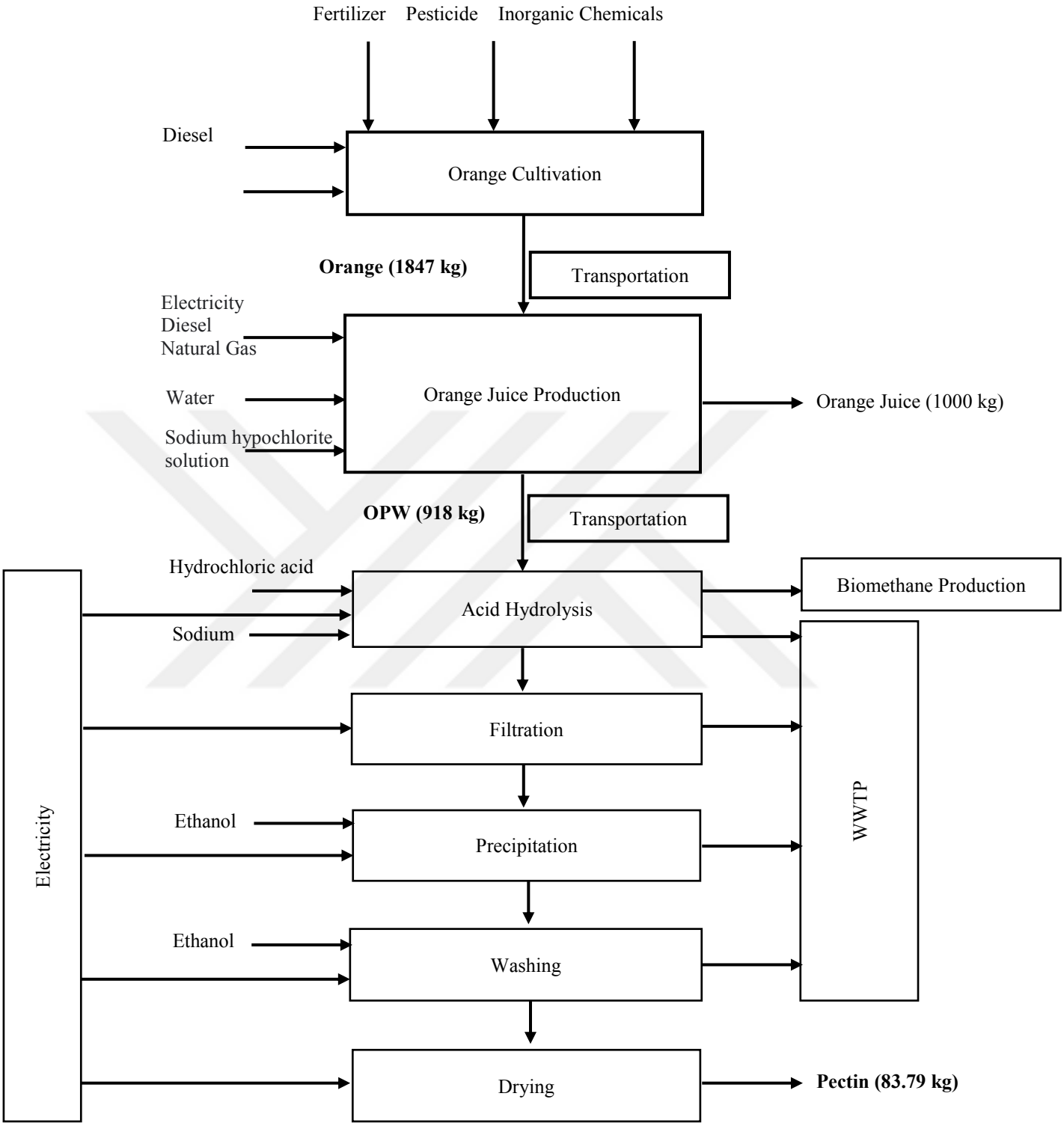


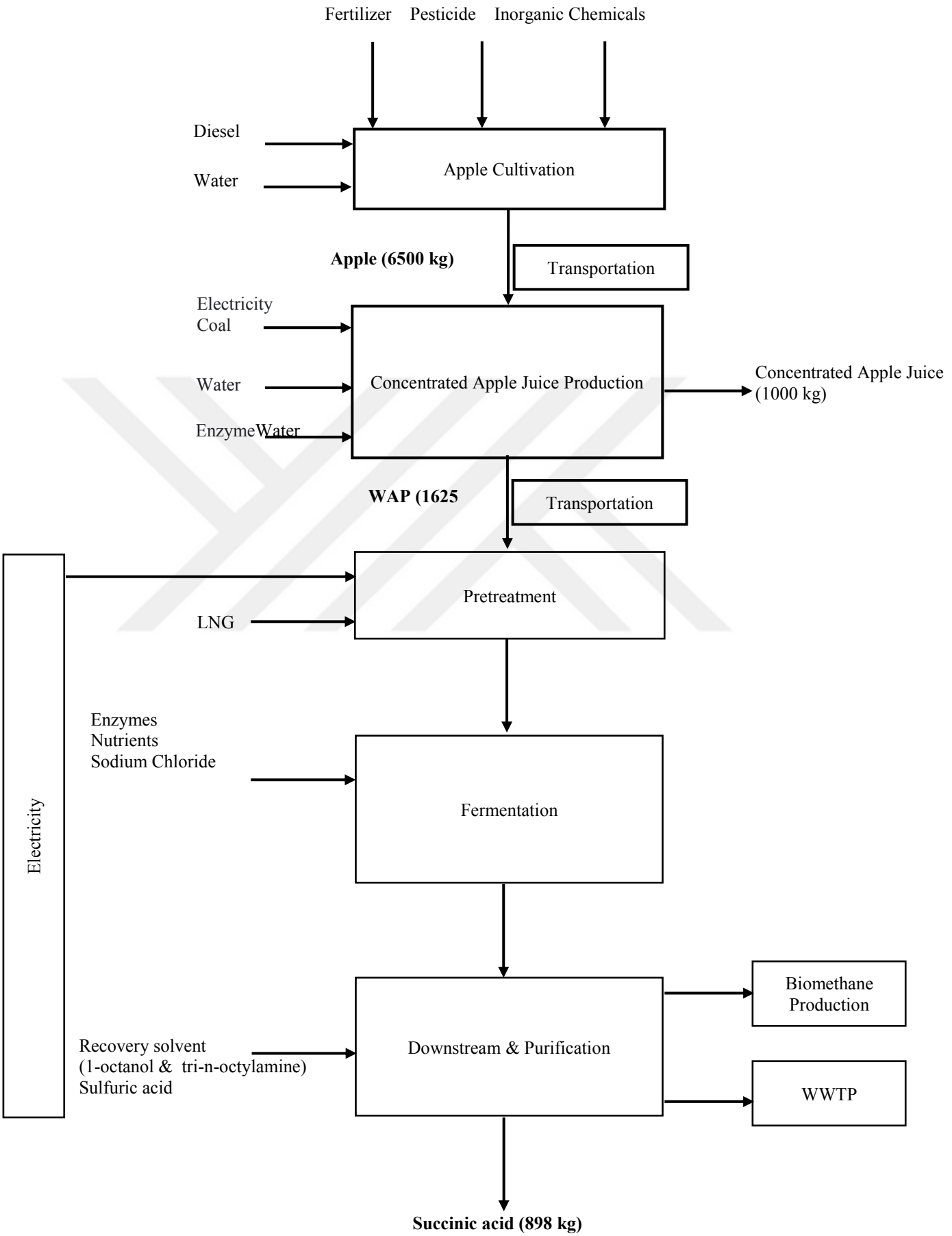
Xylitol Production

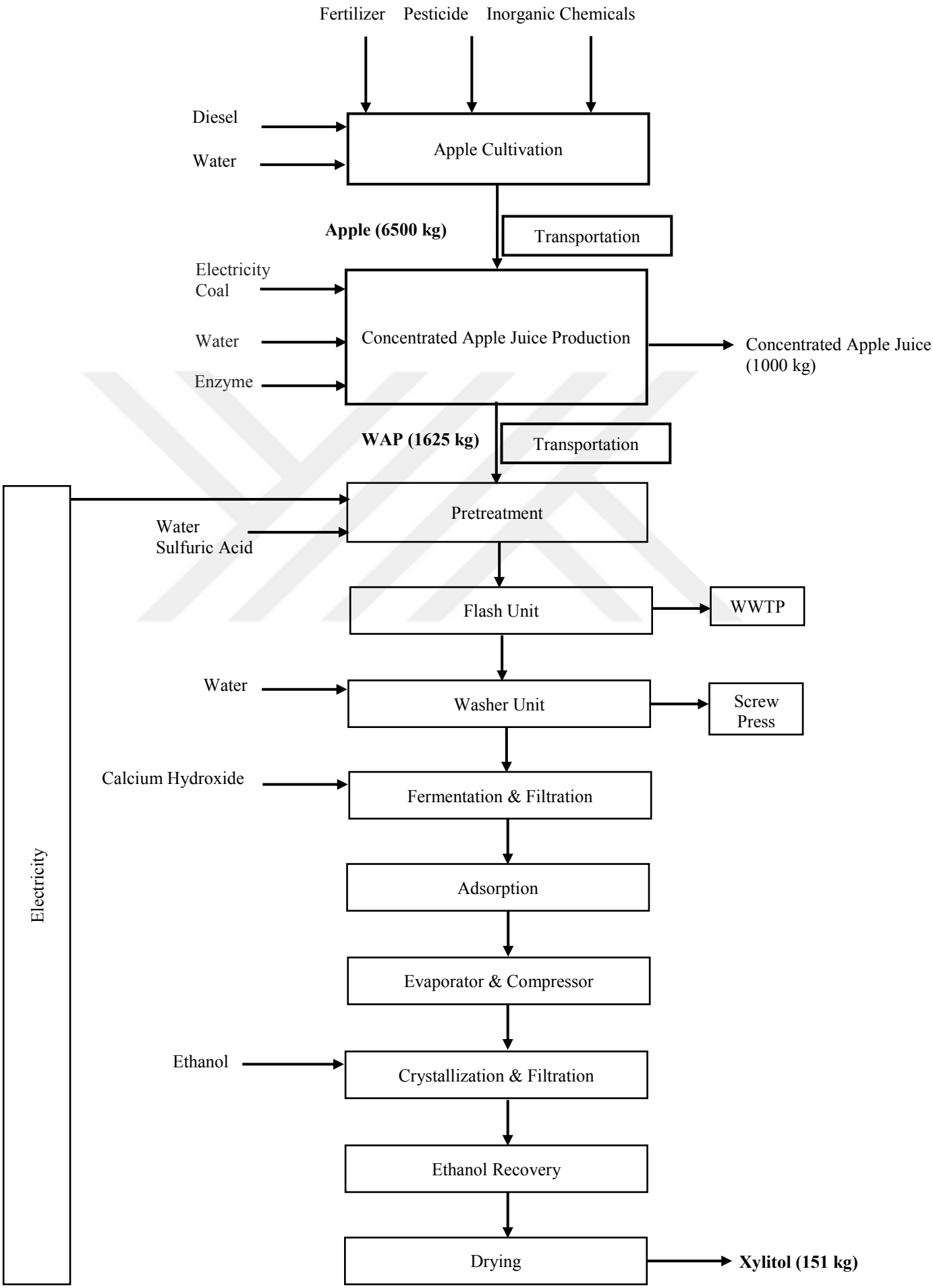


APPENDIX B: SYSTEM BOUNDARIES




Fertilizer
Pesticide
Inorganic Chemicals
Diesel
Orange Cultivation
Orange (1847 kg)
Transportation
Electricity
Diesel
Natural Gas
Water
Sodium hypochlorite solution
Orange Juice Production
Orange Juice (1000 kg)
OPW (918 kg)
Transportation
Hydrochloric acid
Sodium
Acid Hydrolysis
Biomethane Production
Filtration
Ethanol
Precipitation
Ethanol
Washing
Drying
Pectin (83.79 kg)
Electricity
WWTP


Fertilizer
Pesticide
Inorganic Chemicals
Diesel
Water
Apple Cultivation
Apple (6500 kg)
Transportation
Electricity
Coal
Water
Enzyme
Water
Concentrated Apple Juice Production
Concentrated Apple Juice (1000 kg)
WAP (1625
Transportation
Pretreatment
LNG
Electricity
Enzymes
Nutrients
Sodium Chloride
Fermentation
Biomethane Production
Recovery solvent
(1-octanol & tri-n-octylamine)
Sulfuric acid
Downstream & Purification
WWTP
Succinic acid (898 kg)


Fertilizer
Pesticide
Inorganic Chemicals
Diesel
Water
Apple Cultivation
Apple (6500 kg)
Transportation
Electricity
Coal
Water
Enzyme
Concentrated Apple Juice Production
Concentrated Apple Juice (1000 kg)
WAP (1625 kg)
Transportation
Pretreatment
Water
Sulfuric Acid
Flash Unit
WWTP
Water
Washer Unit
Screw Press
Calcium Hydroxide
Fermentation & Filtration
Adsorption
Evaporator & Compressor
Ethanol
Crystallization & Filtration
Ethanol Recovery
Drying
Xylitol (151 kg)
Electricity