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## THE POTENTIAL OF BIOMASS-DERIVED HYDROGEN IN WEST-ANDALUSIA

Deanne van der Slikke<sup>1,2\*</sup>, Karla Guerra<sup>1</sup>, Wiebren de Jong<sup>2</sup>, Pedro Haro<sup>1</sup>

<sup>1</sup>) Bioenergy Group, Chemical and Environmental Engineering Department, Escuela Tecnica Superior de Ingeniería, Universidad de Sevilla, Camino de los Descubrimientos s/n. 41092 Seville, Spain

<sup>2</sup>) Section of Large Scale Energy Storage, Department of Process and Energy, Faculty of Mechanical, Maritime and Materials Engineering, Delft University of Technology, Leeghwaterstraat 39, 2628 CB Delft, The Netherlands

\*Corresponding author: d.f.vanderslikke@student.tudelft.nl

**ABSTRACT:** Hydrogen is believed to be an important energy vector in the transition to a more sustainable future. However, there is a scarcity of relevant studies analysing the realistic potential for hydrogen production in specific European regions. In this study, the potential for biomass-derived hydrogen is comprehensively analysed considering a realistic use of current biogenic residues in relevant sectors. However, not all residues can be used for the production of hydrogen and an analysis has to be made to find how much biomass is suited for hydrogen production, what amounts of hydrogen can be produced from biomass and how these amounts compare to the specific needs for renewable hydrogen in this region. This study focused on hydrogen production from agricultural residues to provide hydrogen for oil upgrading processes in petroleum refineries. The results indicate that around 92% of all agricultural residues in west-Andalusia can be used for hydrogen generation. Gasification and pyrolysis have been identified as the best candidates for the processing of the selected residues. An annual amount of 33,029 tons of biomass-based hydrogen can be used to provide 14% of the hydrogen demand for petroleum upgrading processes, thereby reducing the need natural gas in this industry.

**Keywords:** biomass, hydrogen, residues, integration, gasification.

### 1 INTRODUCTION

Hydrogen is believed to be an important energy vector in the transition to a more sustainable future [5]. Hydrogen storage can facilitate the integration of more renewable energy into the electricity grid, while hydrogen is also used in industry and possibly in other sectors in the near future [17][34]. Hydrogen can be produced from fossil and non-fossil resources [1]. In order to minimize the environmental impact, this study focuses on green hydrogen. Green hydrogen is defined as hydrogen that is produced from renewable energy sources, with the goal of zero to low net carbon emissions and thus minimal environmental impact [14]. Biomass is a renewable energy source that extracts carbon from the air during its growth. However, carbon is emitted during the production of hydrogen from biomass, resulting in zero to low net carbon emissions [31]. Therefore, biomass is considered as green hydrogen source.

The Energy Department of Junta de Andalucía promises the presence of a great biomass wealth in Andalusia [2]. However, not all residues can be used for the production of hydrogen and an analysis has to be made to find how much biomass is suited for the production of hydrogen and what amounts of hydrogen can be produced from biomass. In this study, we will focus on west-Andalusia, that is, the provinces of Huelva, Sevilla and Cádiz. The selected region (west-Andalusia, Spain) has been selected thanks to the presence of a great biomass wealth, relevant industrial presence (two oil refineries), and data availability. This study will explore the potential of biomass-based hydrogen in these three provinces. This will be done by an analysis of the vision of different entities on biomass-based hydrogen, the best suited biomass for the production of hydrogen and the possible applications of hydrogen in west-Andalusia. Lastly, the amount of hydrogen that can be produced from biomass will be compared to the need for hydrogen in west-Andalusia.

### 2 THE NEED FOR RENEWABLE ENERGIES

#### 2.1 History of Andalusia's renewable energy developments

Due to the use of biomass for energy purposes, the average contribution of renewable energies to the primary structure of Andalusia grew rapidly between 2003 and 2006. At that time, biomass represented 80% of the total consumption of renewable energy in Andalusia [25]. The influences of rain and the agricultural season were significant to the contribution of biomass to the primary energy consumption.

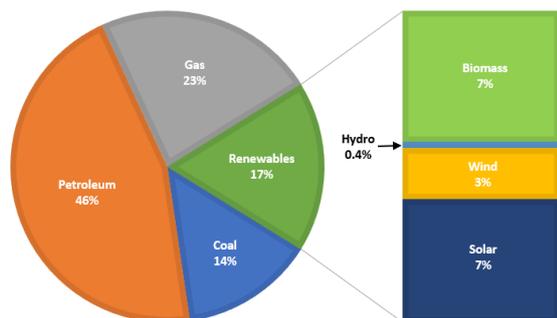
The Andalusian Plan for Energy Sustainability for 2007-2013 (PASENER) [25] contains analyses on the developments on energy sufficiency and renewable energies and urges the public authorities of Andalusia to promote renewable and clean energies. The plan was approved by the Governing Council of Andalusia on November 13, 2007, and this date can be marked as a turning point for the energy model in Andalusia and focuses on demand management, developments in renewable energy and energy efficiency. Even though the plan was made for the period 2007-2013, the region of Andalusia still follows the visions of this plan.

#### 2.2 Renewable resources in Andalusia

The new energy model, as described in the PASENER, emanates from the need to respond to the challenge of climate change and the commitment to guarantee supply, and is based on a greater energy diversity, with high use of native and renewable energy sources. It focuses on the adequate management of a growing demand for energy, the progressive abandonment of fossil fuels in favor of renewable energies, energy saving and efficiency measures and high levels of self-sufficiency. Simultaneously, there is a wish to minimize the impact of instabilities in the energy market.

The abundance of renewable resources in the autonomous region of Andalusia allows the generation of energy from these resources at a high rate. The most important renewable resources in the region are solar, wind and biomass. The distribution of the primary energy

consumption per source is shown in **Figure 1**.



**Figure 1:** Primary energy consumption distribution Andalusia 2018. Based on [4].

With a mainly Mediterranean climate, Andalusia enjoys a high annual average solar radiation of 4.75 kWh/m<sup>2</sup> [25], making it an attractive location for solar photovoltaic and solar thermal energy plants. Also, in the development of these technologies, Andalusia occupies a leading place [2]. Moreover, the abundance of solar energy in the region stimulates growth of certain biomass. The total biomass potential in Andalusia is estimated at 3812 ktoe per year, distributed among several resources, such as agricultural residues, industrial residues, energy crops and forest residues [2].

### 2.3 Andalusia's vision on biomass

There is a wish from the authorities of Andalusia to replace fossil fuels with biomass for energy generation and to promote the use of renewable energies such as biomass, biogas and solar thermal for thermal uses in industry [27]. Moreover, the authorities want to reduce the burning of agricultural residues and promote its use as biomass.

Roughly, two types of biomass can be distinguished. On the one hand, there is biomass that is produced expressly as an energy resource, i.e., energy crops. Production of this type of biomass can revive abandoned areas, but in fact, usually leads to deforestation and occupation of land that would otherwise be used for the production of food crops [28]. On the other hand, there is residual biomass, where waste, often polluting, is transformed into a resource for energy. In view of sustainability and a more circular economy, it is clear that the use of biomass from waste is preferred [2].

### 2.4 Andalusia's vision on hydrogen

In order to achieve decarbonization, energy vectors are needed to incorporate renewable energies into the energy system. Hydrogen is considered to be an important vector in the shift to renewable energies in Andalusia due to its energy storage potential and its potential to serve as an alternative for sustainable transport [5]. Progress needs to be made in storage systems such as hydroelectric pumping, compressed air energy storage and hydrogen storage in salt caverns [2]. The availability of energy storage will play a significant role in Andalusia's potential to become more self-sufficient concerning the supply of energy.

## 3 HYDROGEN APPLICATIONS IN ANDALUSIA

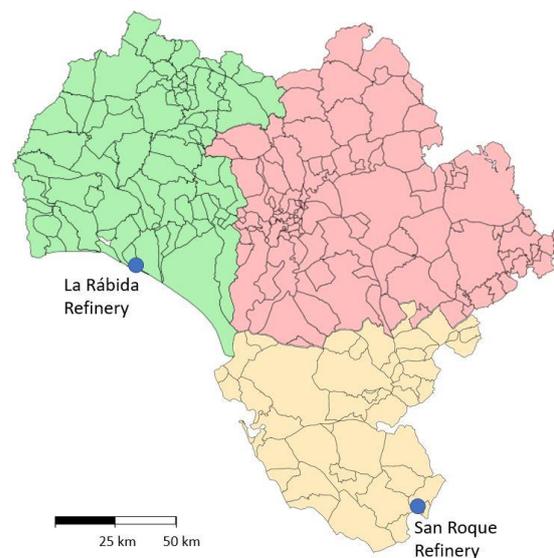
In general, hydrogen can be applied in several

sectors. For instance, as a feedstock for industry, for transport purposes, partially decarbonizing the gas grid by mixing with natural gas and as large scale energy storage for better power system flexibility.

The most promising applications for hydrogen in Andalusia are in hydrogen storage for more flexibility in the power system and to facilitate penetration of renewable energies, and in green hydrogen provision for petroleum refineries. This study only focusses on the last application, green hydrogen for petroleum refineries.

Globally, refineries are the second-largest consumers of industrial hydrogen [34]. Hydrogen is used in the petroleum refinement industry for oil product upgrading in hydrocracking and hydrotreating processes. There has especially been an increase in the demand for hydrogen for hydrocracking, in order to increase the refining yield [21]. High-purity hydrogen (95-99% purity) is required for oil upgrading processes in refineries [12]. It is thought that hydrogen from renewables is essential for the decarbonization of these large industries [20].

West-Andalusia counts two petroleum refineries, La Rábida refinery in the province of Huelva and San Roque refinery in the province of Cádiz, with crude oil throughputs of 9.5 and 12 million tons per year respectively [9][10]. Both are owned by Cepsa and are shown in **Figure 2**.



**Figure 2:** Petroleum refineries in west-Andalusia. Green: Huelva, red: Sevilla, yellow: Cádiz.

The Shell Rheinland Refinery is used as a reference for the hydrogen demand per ton of crude oil. This refinery has a crude oil throughput of 17 million tons per year and a hydrogen demand of 180,000 tons per year [22]. Hence, it is estimated that an average oil refinery requires around 10.6 kg of hydrogen per ton crude oil. This is in accordance with literature, where it is stated that the hydrogen requirement is typically about 1 wt% of crude oil throughput [36]. This results in a total demand of 228,000 tons of hydrogen per year.

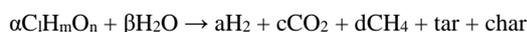
## 4 HYDROGEN PRODUCTION METHODS

Two production technologies are considered for hydrogen production from biomass: biomass gasification and pyrolysis. Each technology has its own advantages

and disadvantages and is able to process different types of feedstock.

#### 4.1 Biomass gasification

In the biomass gasification process, liquid or solid biomass is converted into useful gaseous fuels or chemical feedstock. In our case, biomass is gasified into a fuel gas mixture (syngas), where the interest is in the acquisition of hydrogen. Biomass gasification takes place in a controlled environment and due to its endothermic nature heat is required [40]. A gasifying agent is necessary for the reaction with carbon and hydrocarbons, and the conversion into gaseous products. The main gasifying agents are oxygen or water based [7], where oxygen can be present in its pure form or in the form of air, and water is usually present in the form of steam. It has been shown that using steam as a gasifying agent yields the highest electrical and exergetic efficiencies, and the molar ratio of H<sub>2</sub> in syngas is higher than in processes where oxygen or air is used as a gasifying agent [13]. Biomass steam gasification can be described by the following simplified reaction [35][15]:



Tar is an undesired product that is formed during biomass gasification since it can induce slugging and fouling [15]. Tar formation depends on the type of biomass that is used and can be controlled and minimized with the use of certain catalysts [37].

Supercritical water gasification of biomass is closely related to traditional gasification of biomass and is a promising technology for the production of hydrogen [30]. Water is subjected to its supercritical state and therefore becomes oxidant. Subsequently, biomass is reacted with this supercritical water to produce carbon dioxide and hydrogen. The advantage of this method is that pressurized syngas is created with a higher hydrogen content [30]. Moreover, pre-drying of biomass is not needed since this technology can run with highly wet biomass.

Several factors influence the hydrogen yield of the steam gasification process. The most important ones are biomass type, biomass feed particle size, moisture content, gasification temperature, steam to biomass ratio and catalyst use. In this paper, we will not go into detail on the gasifier parameters, but the influence of biomass type, size and moisture content will be discussed.

The term biomass entails a wide variety of biological organisms that are synthesized through photosynthesis. Biomass refers to plants, animals, microorganisms and the wastes they produce, but it also includes municipal, agricultural and forest waste. Lignocellulosic biomass is mainly composed of cellulose, hemi-cellulose and lignin, but the amount in which they are present depends on the type of biomass. These three components play an important role in the decomposition of biomass. In general, larger amounts of cellulose and lignin yield more gaseous products, which increases the potential of hydrogen recovery [35]. Agricultural wastes, such as olive husks, olive kernels and nut shells contain large amounts of lignin and cellulose and are therefore well suited for gasification [24][38]. Woody forest wastes are also good candidates to be used in gasification.

The size of the biomass particles has a significant effect on hydrogen production. With decreasing particle size, the hydrogen yield and carbon conversion efficiency

increases and the content of char and tar decreases dramatically [29]. This can be explained by the fact that smaller particles have a larger surface area per unit mass, which improves heat and mass transfer, and subsequently improves the gasification reactions [35].

The moisture content of biomass plays an important role in the gasification process. Every kilogram of water in the feed for the gasifier needs around 2242 kJ of energy to vaporize [7] and this energy is lost in the process. Generally, biomass with moisture contents below 35 wt% can be treated in a gasifier, but the best results are obtained for moisture contents of 10-15 wt% [8]. Biomass has typical moisture contents between 30-60 wt%, but for some types it can exceed 90 wt% [7]. Thus, predrying of biomass is needed to reach the highest thermal efficiencies [44]. The remaining moisture evaporates when the feedstock enters the gasifier, thanks to the heat downstream the gasifier.

At Torrgas they believe biomass should not be used directly for the production of syngas due to its variability in quality [11]. The Torrgas process consists of torrefaction of different kinds of biomass and subsequent gasification of the biofuel produced during torrefaction. Scrap wood, agricultural residues and different kinds of waste streams, such as manure and sewage sludge, can be used as feedstock [43]. The biofuel produced during torrefaction can be stored and transported, which replaces the need for storage of biomass. Also, the high-quality biofuel is ten times as energy dense as the original feedstock, requiring less storage space. Torrgas focuses on the production of green methane from syngas, but the produced syngas can be used in different ways, among other things, hydrogen can be obtained from the syngas. A demonstration plant with an input capacity of 1 MWh is currently running in Groningen, the Netherlands, and in the near future a 25 MWh facility will be built. Torrgas focuses on small-to-midscale plants of 5-100 MW.

#### 4.2 Pyrolysis

Pyrolysis is another thermochemical method for the conversion of biomass into hydrogen. This process takes place at elevated temperatures in the absence of oxygen, resulting in gaseous, liquid and solid products [33]:

- The gaseous products consist of H<sub>2</sub>, CH<sub>4</sub>, CO, CO<sub>2</sub> and other gases.
- The liquid products can be used as bio-oil or further processed.
- The solid products are mainly char, pure carbon and other inert materials.

Hydrogen can be produced from biomass through pyrolysis via two routes [33][41]: (1) hydrogen can be obtained directly from the gaseous products when temperatures are high enough and sufficient volatile phase residence time is allowed, or (2) firstly, bio-oil is obtained from biomass through pyrolysis and thereafter, hydrogen can be obtained from bio-oil through steam reforming.

The liquid part obtained in pyrolysis can be separated into a fraction that can be used for hydrogen production and a fraction that can be used for other purposes, such as the production of resins and solvents [41]. Steam reforming and a subsequent water-gas shift reaction process can significantly improve the hydrogen yield [41]. The advantage of this method is that the two steps (pyrolysis and steam reforming) can be carried out

separately and at different places, which is possibly more convenient depending on the biomass availability and transport options [33].

Biomass types characterized by a higher cellulose and hemicellulose content, such as residues from sunflower and olive pruning, have been proven to produce more hydrogen-rich gas than biomass types with a large amount of lignin [47]. However, for the production of hydrogen via the bio-oil route, biomass with a high content of lignin is preferred, since an increase in lignin yields a higher liquid fraction [19]. Woody biomass contains minimal ash and high lignin contents and is therefore well suited for hydrogen production via pyrolysis oil [46].

Clean biomass, with low nitrogen and sulfur contents, is usually preferred for all technologies. However, during pyrolysis, the nitrogen compounds will be transferred to the gaseous phase and sulfur will be mostly present in the char product [18][39]. Therefore, bio-oil from pyrolysis is relatively clean, even when 'dirty' biomass is used as a feed. This makes it possible to use pyrolysis for agricultural wastes that are high in nitrogen and sulfur, since they would contaminate the gasifier if biomass gasification was used.

The pyrolysis process is mainly used for the production of bio-oil from biomass. Therefore, not many examples of pyrolysis plants for the production of hydrogen exist. However, Netteenergy's technology is noteworthy. Netteenergy develops installations that produce valuable products from biomass via pyrolysis. Their technology has been proven to be able to handle wood, rice husks, olive stones and pomace, manure and more [32]. Hydrogen is produced from the syngas by using their SYN2H technology.

## 5 ENERGY STORAGE IN THE BIOMASS-TO-HYDROGEN PROCESS

Storage of energy in the form of hydrogen is one method to ensure the match between power generation and demand, but the amount of available hydrogen from biomass fluctuates seasonally. Energy can be stored at different stages of the biomass-to-hydrogen process. Storage of biomass has the advantage that hydrogen can be produced more flexibly and the obtained biomass does not have to be converted into hydrogen directly, which can be advantageous in periods when a lot of biomass is available. The disadvantages of storage of biomass are the related costs and logistics. Pyrolysis oil has the advantage that it is easily transported and stored. However, gasification is preferred for some types of biomass, therefore storage of pyrolysis oil is not always an option. Storage of hydrogen itself has the advantages that it can be stored on large scale and can be directly converted into energy when needed. However, some hydrogen storage technologies are more energy intensive than others, thus special attention will need to be paid to choosing the right hydrogen storage technology.

## 6 BIOMASS IN WEST-ANDALUSIA

Andalusia knows a large wealth when it comes down to biomass [26]. An evaluation of all residues in Andalusia by the Energy Department of Junta de Andalucía shows that there is a potential of almost 8

million tons of biomass for the production of energy in west-Andalusia alone [3]. The available biomass in west-Andalusia has been grouped as follows: agricultural residues, forestry residues, industrial residues, urban residues, animal residues and energetic crops. This study only focusses on the first group. The characteristic of the different types of biomass, their availability, best suited conversion technologies and hydrogen yields have been evaluated.

### 6.1 Biomass source selection

Agricultural residues include all waste that is generated directly on the farmland. Agricultural residues can be divided in two groups: woody residues from the pruning of fruit, citrus, vine and olive trees; and herbaceous residues that remain after harvest. The crops with the greatest potential or availability of biomass are those that [26]:

- Produce a large amount of biomass per unit area.
- Do not have alternative uses of great economic viability, such as animal feed.
- Are easily and economically obtainable from the farmland.
- 

The Energy Department of Junta de Andalucía has made an assessment of the potential of biomass in its region [3][26]. The energy density (LHV) and amount of annually available agricultural residues in west-Andalusia can be found in **Table I**.

**Table I:** Overview of agricultural residues

Biomass type	Available biomass [tons]	LHV dry basis [MJ/kg]	N dry basis [wt%]	S dry basis [wt%]
Citrus	36,493	-	-	-
Fruit	6,128	-	-	-
Vine	25,647	-	-	-
Olive grove	296,869	17.63	0.59	0.05
Tomato	25,396	-	-	-
Greenhouse	35,763	-	-	-
Cotton	589,515	16.2	1.24	0.95
Sunflower	375,577	16.1	1.2	0.1
Rice	145,910	13.95	0.87	0.18

A criteria that has been used to determine whether an agricultural residue is fit for the production of hydrogen is the amount of annually available biomass. The minimum amount has been set at 50,000 tons of available biomass per year. This eliminates the use of citrus, fruit, vine, tomato and greenhouse residues.

Olive grove, cotton, sunflower and rice residues are left as potential agricultural energy sources for the production of hydrogen. These four types of biomass have a high energy density and are available in large amounts. Gasification is the preferred technology, because of its development, efficiency and scale. Therefore, olive grove, sunflower and rice residues will be best handled by a gasifier. However, cotton will be converted into hydrogen via pyrolysis due to its high sulfur content.

Olive groves stand out as a biomass among all others. It is one of the most precious products of Andalusia and is a source of numerous by-products. Olive grove pruning

happens yearly for table olive trees and every two years for olive oil trees. These wastes were traditionally linked to the use as firewood. However, lately the use of olive pruning splinters for electricity generation has experienced a significant increase due to an improvement in profitability [26]. The need to introduce new biomass sources in existing electricity generation plants led to many power generators to gain interest in wood chips.

### 6.2 Hydrogen yield determination

Experiments on gasification of olive tree pruning have been performed and showed a potential hydrogen yield of 36.6 g H<sub>2</sub>/kg pruning [45].

Cotton residues contain large amounts of sulfur and nitrogen [42], which will damage the gasifier. For this reason, pyrolysis would be best suited to treat cotton residues for the production of hydrogen. Pyrolysis of cotton stalks can reach hydrogen yields of 13.6 g H<sub>2</sub>/kg of biomass [6].

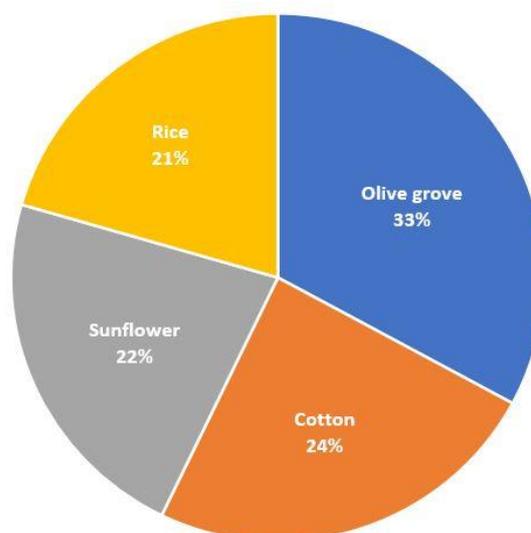
Sunflower residues, mainly sunflower stalks, can be gasified as well. However, no relevant literature provided the hydrogen yield for gasification of sunflower residues. Therefore, the possible hydrogen yield from pyrolysis of sunflower residues will be used as a reference, which can reach a hydrogen yield of 9.71 mol H<sub>2</sub>/kg of biomass [16], or 19.6 g H<sub>2</sub>/kg of biomass.

After harvesting of rice, mainly rice straw is left. A lignocellulosic with, up to now, no valuable destination. Gasification of rice straw can reach hydrogen yields of up to 23.07 mol H<sub>2</sub>/kg of biomass [23], which is equal to 46.5 g H<sub>2</sub>/kg of biomass.

**Table II:** Overview of hydrogen yield per biomass source

Biomass source	Available biomass [tons]	Hydrogen yield [g H <sub>2</sub> /kg biomass]	Annual hydrogen yield [tons]
Olive	296,869	36.6	10,865.4
Cotton	589,515	13.6	8,017.4
Sunflower	375,577	19.6	7,361.3
Rice	145,910	46.5	6,785.8
Total	1,407,871		33,028.9

**Table II** provides an overview of the hydrogen yield per biomass source and the total annual hydrogen yield for the selected agricultural residues. A total amount of more than 1.4 million tons of agricultural residues per year is considered as a potential source for the production of hydrogen in west-Andalusia, this is around 18% of the total amount of biomass available in west-Andalusia according to the Energy Department of Junta de Andalucía [3]. All other biomass sources are not considered in this study and are available for use elsewhere. The distribution of the contribution to the production of hydrogen per type of biomass for west-Andalusia is shown in **Figure 3**.

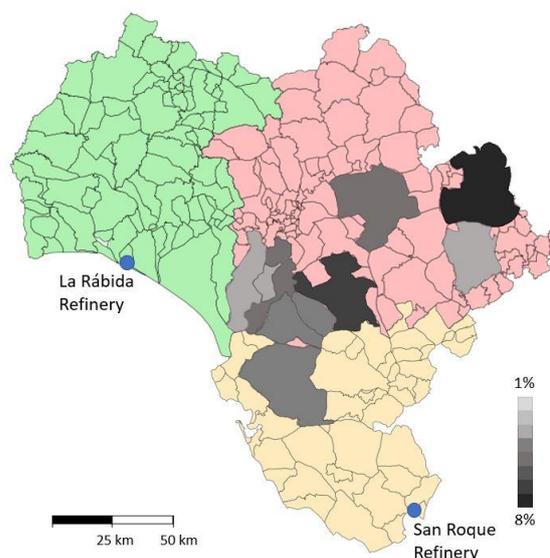


**Figure 3:** Distribution of the hydrogen provision per biomass type.

## 7 HYDROGEN PROVISION IN WEST-ANDALUSIA

By gasification and pyrolysis of the selected agricultural residues 33,029 tons of hydrogen can be obtained per year. This means that 14% of the hydrogen demand by petroleum refineries in west-Andalusia can be provided as green hydrogen obtained from biomass.

**Figure 4** shows the ten municipalities with the highest production of agricultural residues in west-Andalusia. The percentage of agricultural residue in the respective municipality over the total amount of agricultural residue has been indicated by the grey-scale. Data has been obtained and adjusted from a publication by the Energy Department of Junta de Andalucía [3]. It can be seen that most biomass originates from the province of Seville, while the refineries are located in the provinces of Huelva and Cádiz. Transportation and storage of biomass, pyrolysis oil or hydrogen will thus have to be taken into account.



**Figure 4:** Origin of agricultural residues and the refinery locations. Green: Huelva, red: Sevilla, yellow: Cádiz.

## 8 CONCLUSION

The application of green hydrogen for petroleum refineries in west-Andalusia was selected in this study. Selected agricultural residues will be used for the production of hydrogen for oil upgrading processes in refineries. Gasification and pyrolysis of these residues are considered as the best suited technologies to produce hydrogen from olive grove, cotton, sunflower and rice pruning residues. The annual hydrogen yield has been calculated to be 33,029 tons, thereby being able to provide 14% of the annual required amount of hydrogen by oil refineries in west-Andalusia and thus reducing the need for natural gas for hydrogen production in this industry.

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