

# Life cycle greenhouse gas emissions of hydrogen fuel production from chlor-alkali processes in the United States

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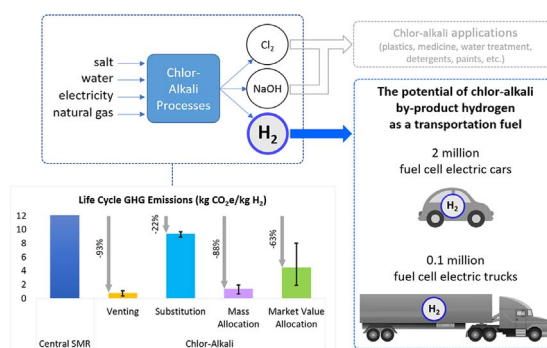
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## HIGHLIGHTS

- By-product H<sub>2</sub> production is compared to the conventional steam methane reforming pathway.
- Hydrogen from chlor-alkali processes provides life-cycle greenhouse gas emissions reductions.
- Detailed plant-by-plant and regional analyses are conducted.
- The impact of combined heat and power for the chlor-alkali industry is evaluated.
- Different co-product treatment methods are compared.

## GRAPHICAL ABSTRACT



## ARTICLE INFO

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## ABSTRACT

By-product hydrogen from chlor-alkali processes can help meet the increasing demand for hydrogen fuel in early fuel cell electric vehicle markets (e.g., California) in the U.S. Hydrogen produced from chlor-alkali plants is typically combusted for process heat on site, vented to the atmosphere (i.e., wasted), or sold to the external merchant hydrogen market. Whether it is combusted, vented, or sold as a commodity, relevant information is lacking as to the life-cycle environmental benefits or trade-offs of using by-product hydrogen from chlor-alkali plants. A life-cycle analysis framework was employed to evaluate well-to-gate greenhouse gas (GHG) emissions associated with by-product hydrogen from chlor-alkali processes in comparison with hydrogen from the conventional centralized natural gas steam methane reforming (central SMR) pathway. U.S.-specific, plant-by-plant, and up-to-date chlor-alkali production characteristics were incorporated into the analysis. In addition to the venting and combustion scenarios, to deal with the multi-functionality of the chlor-alkali processes that simultaneously produce chlorine, sodium hydroxide, and hydrogen, two different co-product allocation strategies were adopted—mass allocation and market value allocation. It was estimated that by-product hydrogen production from chlor-alkali processes creates 1.3–9.8 kg CO<sub>2</sub>e/kg H<sub>2</sub> of life-cycle GHG emissions on average, which is 20–90% less than the conventional central SMR pathway. The results vary with co-product treatment scenarios, regional electric grid characteristics, on-site power generation, product prices, and hydrogen yield. Despite the variations in the results, it was concluded that the life-cycle GHG emission reduction benefits of using by-product hydrogen from chlor-alkali processes are robust. With a diverse set of scenario analyses, the study developed a comprehensive and detailed life-cycle GHG emissions inventory of the chlor-alkali by-product hydrogen pathway and quantified sensitivity indices in the context of different assumptions and input parameter values.

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## 1. Introduction

Hydrogen is a clean fuel, creating only water and no harmful exhaust emissions at the point of use. It can play an important role in transitioning toward sustainable energy systems across sectors, including transportation, buildings, and power. The overall environmental footprint (e.g., carbon emissions) of hydrogen energy, however, depends on the source of hydrogen (fossil fuels, nuclear, renewable sources, etc.), the hydrogen production process (steam methane reforming [SMR], water electrolysis, etc.), and the source of energy used for hydrogen production, storage, and delivery [1]. Efficient and clean sources of hydrogen, promoting diversity and flexibility, are needed [2]. In fact, the search for a cleaner source of hydrogen fuel, such as wind energy, is an active research area [3,4]. The interaction and integration of hydrogen energy systems and applications with other components (e.g., the electric grid) is also a topic of research interest [5,6]. Although diverse research is ongoing on hydrogen energy systems, most of the recent discussions have centered on hydrogen fuel cell electric vehicles (FCEVs), as the world's first mass-produced FCEVs (e.g., Hyundai Tucson, Honda Clarity, and Toyota Mirai) have recently been introduced in various automotive markets.

In spite of some barriers (e.g., refueling infrastructure availability, overall cost), the demand for hydrogen as a transportation fuel is expected to grow in the years to come, with an increasing number of hydrogen FCEVs on the road. In the U.S., California and eight other zero emission vehicle (ZEV) states have plans to deploy a large number of zero (tail-pipe) emission vehicles, including FCEVs. In California, for example, owing to the ZEV mandate and other incentives, around 40,000 FCEVs could be on the road by 2025 [7,8]. It is uncertain exactly how many FCEVs will be deployed by when and where. However, as future FCEV adoption unfolds, it is critical to ensure that the hydrogen fuel powering those FCEVs is produced with the lowest possible cost and environmental impacts [9]. Sourcing hydrogen from less-polluting processes not only helps protect the environment but also brings tangible economic benefits by leveraging current incentive programs for ZEVs. For instance, with the low-carbon fuel standard (LCFS) in California [10], hydrogen fuel from centralized natural gas SMR (or central SMR) plants generates credits in the range of \$1.2–\$1.6/kg H<sub>2</sub> for substituting petroleum gasoline, at the average traded price of approximately \$100/tonne CO<sub>2</sub>e in 2016 and 2017. That being said, by-product hydrogen, simultaneously produced with other main products (e.g., chlorine, ethylene) from industrial processes, which is sometimes vented (wasted) to the atmosphere, presents a very interesting case. Such by-product hydrogen is already produced at scale and at a low cost (about \$1/kg H<sub>2</sub>), requires no new capital investment because of its high purity (> 99%), and thus can serve emerging FCEV markets [11,12]. An FCEV consumes about 200 kg of hydrogen fuel per year, assuming 12,000 miles of annual travel distance per vehicle [13] and a fuel economy of 65 miles/kg H<sub>2</sub> [14]. If by-product hydrogen (from industrial sources) is not vented/wasted but rather used as a fuel for, say, 10,000 FCEVs, annual revenue from LCFS credits could be on the order of a few million U.S. dollars, depending on the life-cycle greenhouse gas (GHG) emissions and the CO<sub>2</sub> credit price under the LCFS. The exact environmental and associated monetary benefits, however, will depend on the overall emissions (if any) that are created over the life cycle of the by-product hydrogen, for which this study provides a detailed and comprehensive analysis.

Among various industrial sources of by-product hydrogen, the chlor-alkali industry is considered one of the most promising low-cost hydrogen sources in the near term [11,12]. Chlor-alkali processes produce hydrogen as a by-product, along with chlorine and sodium hydroxide as main co-products, via the electrolysis of aqueous sodium chloride. The reported annual hydrogen production capacity of the U.S. chlor-alkali industry is about 0.4 million tonne [15]. If fully dedicated for transportation fuel, by-product hydrogen from chlor-alkali processes can power more than 2 million FCEVs. Despite such potential as a large-

scale hydrogen fuel source, a detailed and up-to-date life-cycle environmental benefit analysis of by-product hydrogen from the U.S. chlor-alkali industry is not available. ACC-Franklin [16] developed a life-cycle inventory for chlorine and sodium hydroxide production from chlor-alkali processes in the U.S. However, the inventory lacks hydrogen-specific information and is based on data collected in the early 1990s and 2003 for a small sample of chlor-alkali plants and on national average emissions factors for electricity generation. Electricity is a major energy input for chlor-alkali processes. That being said, temporal evolution, as well as regional characteristics of the U.S. chlor-alkali industry and the electric grid, were not properly accounted for in previous studies. As a matter of fact, both the chlor-alkali industry and the electric grid in the U.S. have undergone a remarkable transformation in recent years. For the chlor-alkali industry, there has been a drastic change in the production share of the three major electrolysis cell technologies—diaphragm, membrane, and mercury [17]. Between 2005 and 2015, the capacity share of membrane cells increased by 94%, while the share of diaphragm and mercury cells decreased by 20% and 90%, respectively. For the electric grid, natural gas (34%) surpassed coal (30%) as the biggest fuel source for electricity generation in 2016, representing a substantial change from the 20% natural gas and 50% coal share in 2006 [18,19]. Also, the electricity generation share of wind power rose by 0.5% annually over the last decade, reaching the same generation share as hydroelectric power in 2016 (6.5%, or 266 TWh/year) [19,20]. Given such significant technological and market transformations over the past decade, a more up-to-date and detailed life-cycle inventory is needed for hydrogen production from chlor-alkali processes in the U.S.

A relatively large number of chlor-alkali studies exist for other countries. Euro Chlor [21] assessed the European chlor-alkali industry's average life-cycle environmental performance for reference year 2011. In another European case study, Jung et al. [22] compared two electrolysis technologies (membrane vs. oxygen depolarized cathode) for chlorine and sodium hydroxide production along with their life-cycle environmental performance. Garcia-Herrero et al. [23] evaluated four different chlor-alkali electrolysis cell types and their life-cycle environmental impacts in Europe. It is important to acknowledge, however, that the European and U.S. chlor-alkali industries have distinctly different characteristics. For example, membrane cell is the dominant electrolysis technology in Europe, accounting for 66% of production capacity, followed by mercury (17%) and diaphragm (17%) [24,25]. In the U.S., on the other hand, diaphragm technology makes up 55% of total chlor-alkali production capacity, and membrane 45% [17]. Since different electrolysis cell technologies have different energy demand characteristics in terms of composition and intensity [24,26], it is critical to account for the differences in the chlor-alkali industries in various countries or regions. Similarly, different electricity generation characteristics between Europe and the U.S., and the regional variations within the U.S., highlight the need for a U.S.-specific life-cycle inventory with a focus on by-product hydrogen production from chlor-alkali plants.

Above all, the current body of literature does not provide relevant information on hydrogen fuel production, nor is it based on a plant-by-plant regional analysis for the U.S. In chlor-alkali processes, unlike chlorine or sodium hydroxide, hydrogen is not a main product; it is a by-product. Chlor-alkali plants are built to produce chlorine and sodium hydroxide, not hydrogen. Hydrogen, as a by-product, produced along with chlorine and sodium hydroxide, may be sold, burned on site for process heat, or vented; each option requires a careful selection of a system boundary for estimating the corresponding environmental footprint. Hydrogen also requires a different set of sub-processes from those for chlorine or sodium hydroxide. This fact complicates the allocation of environmental burdens between the main products (chlorine and sodium hydroxide) and the by-product (hydrogen). Therefore, shifting the focus to evaluating the environmental burden of hydrogen production creates numerous challenges for life-cycle analysis (LCA),

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