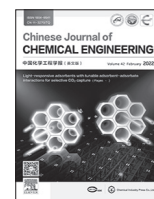




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Full Length Article

Life cycle assessment of HFC-134a production by calcium carbide acetylene route in China

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ABSTRACT

HFC-134a is a widely used environment-friendly refrigerant. At present, China is the largest producer of HFC-134a in the world. The production of HFC-134a in China mainly adopts the calcium carbide acetylene route. However, the production route has high resource and energy consumption and large waste emission, and few of the studies addressed on the environmental performance of its production process. This study quantified the environmental performance of HFC-134a production by calcium carbide route via carrying out a life cycle assessment (LCA) using the CML 2001 method. And uncertainty analysis by Monte-Carlo simulation was also carried out. The results showed that electricity had the most impact on the environment, followed by steam, hydrogen fluoride and chlorine, and the impact of direct CO₂ emissions in calcium carbide production stage on the global warming effect also could not be ignored. Therefore, the clean energy (e.g., wind, solar, biomass, and natural gas) was used to replace coal-based electricity and coal-fired steam in this study, showing considerable environmental benefits. At the same time, the use of advanced production technologies could also improve environmental benefits, and the environmental impact of the global warming category could be reduced by 4.1% via using CO₂ capture and purification technology. The Chinese database of HFC-134a production established in this study provides convenience for the relevant study of scholars. For the production of HFC-134a, this study helps to better identify the specific environmental hotspots and proposes useful ways to improve the environmental benefits.

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1. Introduction

HFC-134a (1,1,1,2-tetrafluoroethane, CH₂FCF₃) is a widely used environment-friendly refrigerant, which has become a very effective and safe alternative for R12 because of its excellent comprehensive performance [1]. It is mainly used in automobile air conditioners, as well as in central air conditioners, commercial refrigeration, ice cream machines and other refrigeration equipments. At the same time, it can be applied to aerosol propellant, medical aerosol, polymer physical foaming agent, and magnesium alloy shielding gas and other fields. At present, global warming has

caused wide public concern. Global warming potential (GWP) is an index based on the radiation characteristics of well-mixed greenhouse gases. It is commonly used metric for comparing the impact of various greenhouse gases. The GWP of HFC-134a is as high as 1300 [2]. The European Union (EU) had issued clear automotive air conditioning refrigerant management policies [3], which bans the use of refrigerant with a GWP of more than 150 in all automobile air conditioning systems sold from 2017. R450A has a lower GWP (549) [4] and has been included in the United States (US) Significant New Alternatives Program (SNAP) by the US Environmental Protection Agency (EPA) as a HFC-134a alternative. It is a mixture of 42% HFC-134a and 58% HFO-1234ze by mass, and has been used in a large number of refrigeration and air conditioning equipments. At present, China has not yet controlled the use of HFC-134a. The production capacity of HFC-134a in China was

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about 243 kilotons in 2016, increasing to 303 kilotons in 2019 [5,6]. Due to the gradual reduction of HFC-134a production capacity in developed countries, China has become the largest producer of HFC-134a in the world. The export volume reached 106 kilotons in 2019 [5], and export has become an important channel for HFC-134a production digestion. The production process of HFC-134a mainly include calcium carbide acetylene route and petroleum ethylene route. Its main raw material is trichloroethylene, while the main raw material of trichloroethylene is acetylene or ethylene. Due to the abundant coal reserves and low petroleum and natural gas reserves in China, trichloroethylene is basically produced from calcium carbide acetylene as raw material. However, compared with the foreign petroleum ethylene route, the production of HFC-134a by calcium carbide acetylene route in China has higher resource and energy consumption and large waste emission.

LCA is the compilation and evaluation of the inputs, outputs and the potential environmental impacts of a product system throughout its life cycle. It is one of the most scientific and effective methods for assessing environmental impact recommended by many scholars and associations [7]. It can identify the key points and sources of serious pollution during production, and provide constructive suggestions for reducing the environmental burden. The current researches on HFC-134a mainly focused on its physical properties [8,9], the performance of HFC-134a air conditioning systems [2,10] and their impacts on global warming [11,12]. Bovea *et al.* [13] compared the environment impacts between HFC-134a and other refrigerants in the refrigeration systems *via* LCA method. However, few of the studies addressed on quantifying the impact of HFC-134a production process on the environment. McCulloch and Lindley [14] researched the resources and energy consumption and pollutant emissions required in the production process of HFC-134a by the life cycle inventory (LCI) analysis approach, but did not quantify its impact on the environment, and did not optimize the production process.

In this area, previous studies have been limited to the environmental impact of HFC-134a as a refrigerant for automobiles, with little attention to the environmental problems caused by the production of HFC-134a. One of the main reasons is that HFC-134a production process is so extensive that it is difficult to collect accurate and detailed data on various aspects required for environmental assessment. Through field research, cooperation with manufacturers, online information search and data analysis and processing, we have overcome various difficulties one by one.

This study focused on the “cradle to gate” LCA of HFC-134a production by calcium carbide route. A LCA model was constructed, and the CML 2001 method was selected to quantitatively evaluate the environmental burden of HFC-134a production. Monte-Carlo simulation was used to evaluate the variation range of the results. This study established a Chinese database of HFC-134a production, identified the key points and raw and auxiliary materials with great environmental impact in the production process, and provided an effective basis for energy saving and emission reduction.

2. Materials and Methods

2.1. Goal and scope definition

This study aims to build a complete life cycle inventory of HFC-134a production, quantify the environmental burden, identify critical substances during production, and provide valuable suggestions for the reduction of environmental burden. Functional unit provides a quantified reference for the relevant inputs and outputs

of an investigated system [15]. In this study, 1 ton HFC-134a production was selected as the functional unit.

System boundary (Fig. 1) was established by the cradle-to-gate approach, which mainly included raw materials and energy production, materials transportation, and waste disposal associated with HFC-134a production. HFC-134a production included three process stages: calcium carbide production, trichloroethylene production and HFC-134a production.

Calcium carbide production: Lime and coke were mixed and added into the closed calcium carbide furnace. They reacted under the high temperature (2000–2200 °C) generated by electric arc and converted into calcium carbide, and produced a large amount of furnace gas was, in which the CO content was about 82%–90%. The reaction equations were as follows:



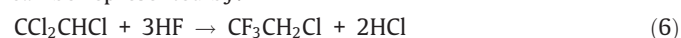
Refer to the literature for a detailed description of the process [16]. The furnace gas was treated with dry purification technology and then sent to the lime kiln as fuel for calcining limestone. A large amount of CO₂ emitted from furnace gas combustion and limestone calcination. The carbon-containing solid waste and lime dust were pressed into pellets, then burned as fuel and reused in the calcium carbide furnace respectively. Other solid waste was used as raw materials for cement. The main inputs at this stage included electricity, limestone, coke, electrode paste and water, and the main outputs included calcium carbide, direct emissions (SO₂, NO_x, CO₂) and solid waste.

Trichloroethylene production: Calcium carbide hydrolyzed with water to produce acetylene which reacted with chlorine gas to produce tetrachloroethane after purifying and cooling. Then tetrachloroethane was dehydrochlorinated to produce trichloroethylene. The reaction equations were as follows:



The catalyst was recycled and processed by a specialized company, so its impact was not considered. The main inputs in this process included calcium carbide, chlorine, utilities (electricity, steam, water, nitrogen and compressed air), sodium hydroxide, ferric chloride and coal. The outputs included five products (trichloroethylene, tetrachloroethylene, hydrogen chloride, high-boiling product and low-boiling product), direct emissions (Cl₂, HCl, trichloroethylene and NMVOC) and solid waste. Since trichloroethylene production is a multi-product output system, it is necessary to allocate the inputs and outputs to different products. According to International Standardization Organization (ISO) recommended allocation order, priority was given to physical attributes (such as mass), followed by economic value [17]. At this stage, mass allocation was adopted for the sake of fairness. The allocation factors of trichloroethylene, tetrachloroethylene, hydrogen chloride, high-boiling product and low-boiling product were 73.8%, 2.2%, 20.1%, 2.4% and 1.5%, respectively. After allocation, trichloroethylene was used for downstream production, while the use of other four products was not considered in this study.

HFC-134a production: The reactions mainly involved in the HFC-134a production were completed in two stages on the chromium-based catalyst at elevated temperature. The processes can be represented by:



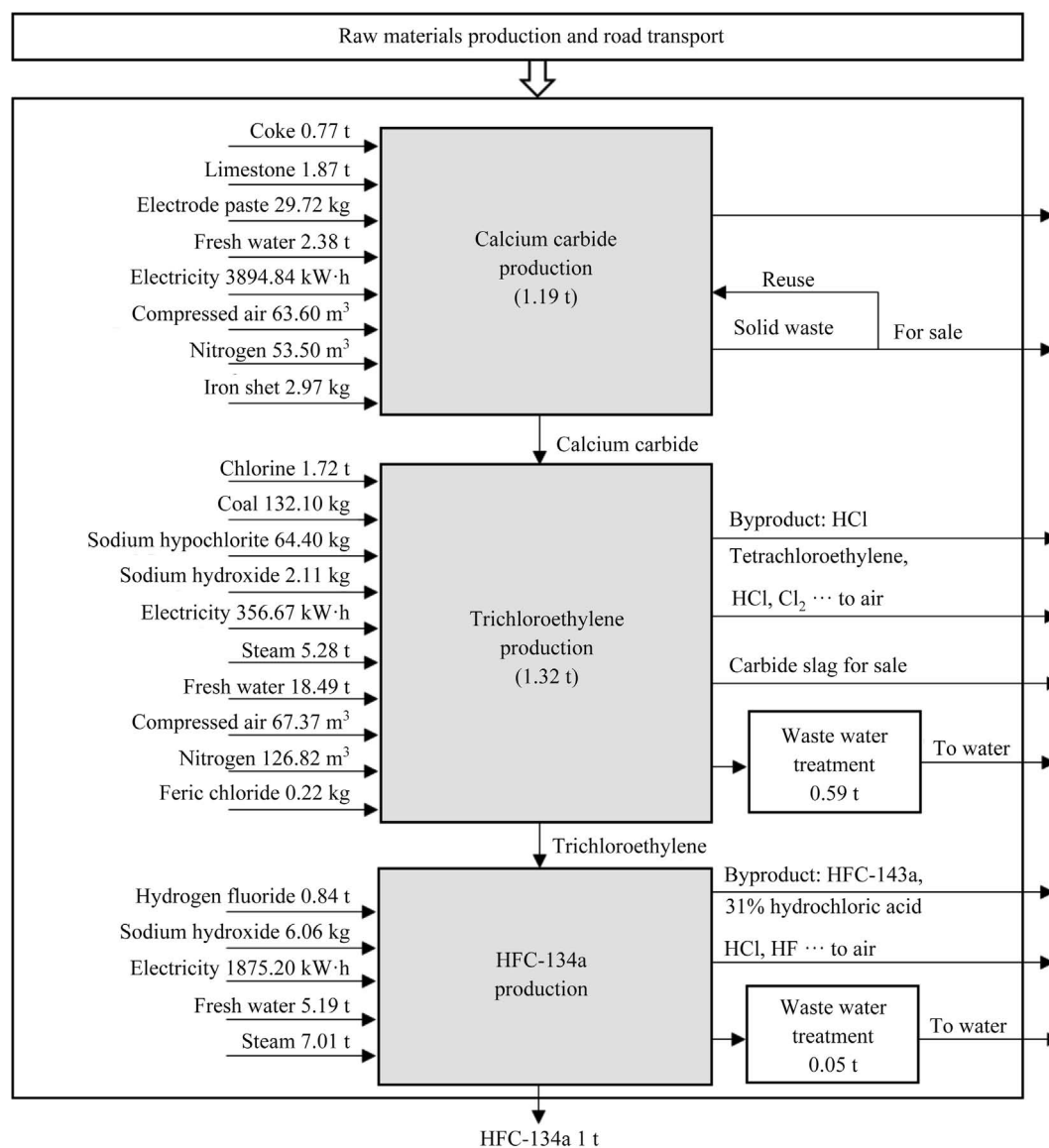


Fig. 1. System boundary of HFC-134a production by calcium carbide acetylene route.



Trichloroethylene was converted to HCFC-133a ($\text{CF}_3\text{CH}_2\text{Cl}$) in the first stage reactor with a conversion rate of more than 99%. Then all materials entered the second stage reactor, where HCFC-133a reacted with anhydrous hydrofluoric acid to produce HFC-134a. It is much more difficult to substitute the last remaining chlorine substituent with fluorine at this stage. The reaction equilibrium is biased towards the starting materials, so hydrogen chloride needs to be removed, while a large amount of other reagents are circulating in the reaction system. The whole process was enclosed. The intermediate reagents were physically retained in the equipment and are unlikely to be released into the atmosphere. The effect of catalyst was also not considered at this stage. The main inputs included trichloroethylene, anhydrous hydrogen fluoride, sodium hydroxide and utilities (electricity, steam, water), and the outputs included three products (HFC-134a, 31% hydrochloric acid, HFC-143a) and direct emissions (HF, HCl, trichloroethylene and NMVOC). Since HFC-134a production was also a multi-product system, an allocation was conducted. In the case of mass allocation, the allocation factors of HFC-134a, HFC-143a and 31%

hydrochloric acid (calculated based on the net content of hydrogen chloride) were 50.1%, 0.2% and 49.7%, respectively. The economic value of 31% hydrochloric acid was far lower than that of HFC-134a, but it carried nearly half of the environmental burden, which was not in line with the production goal of the factory. Therefore, economic value allocation was adopted in this study. HFC-134a carried most of the environmental burden with an allocation factor of 95.1%. The allocation factors of HFC-143a and 31% hydrochloric acid were 0.5% and 4.4%, respectively.

2.2. Life cycle inventory

Raw data on raw materials, energy consumption and direct emissions were obtained from the 2017 annual reports of three typical Chinese producers. The production data of calcium carbide and trichloroethylene were from plants in Inner Mongolia Autonomous Region, with an annual output of 600 kilotons of calcium carbide and 40 kilotons of trichloroethylene. And the data of HFC-134a production was from plants in Shaanxi Province with an annual output of 12 kilotons of HFC-134a. The life cycle inventory of HFC-134a production by the calcium carbide acetylene route

was shown in Supplementary Material S1. Background data (e.g., electricity, steam, water, coal, chlorine, hydrogen fluoride, and sodium hydroxide, etc.) were obtained from the GaBi database included in the GaBi software [18]. Coal-based electricity and coal-fired steam were considered in this study, as coal played a leading role in China's energy production.

The main transportation materials involved in this study were limestone, coke, calcium carbide, chlorine, trichloroethylene and hydrogen fluoride. The calcium carbide and trichloroethylene production plants are located in Inner Mongolia Autonomous Region, and the two plants are adjacent. Limestone and chlorine were from the surrounding areas, coke was from Shaanxi Province, and hydrogen fluoride was from Hubei Province. The transportation distance was calculated according to the actual distance, so the transportation distances of limestone, coke, calcium carbide, chlorine, trichloroethylene and hydrogen fluoride were 50 km, 250 km, 3 km, 3 km, 802 km and 668 km respectively. All transportation modes were trucks.

2.3. Life cycle impact assessment (LCIA) method

In this study, LCA models were established by GaBi software and GaBi databases. CML 2001 method was adopted in LCA evaluation [19], which was recognized as one of the most world-widely used midpoint methodology. The investigated impact categories included abiotic depletion potential (ADP elements), abiotic depletion potential (ADP fossil), acidification potential (AP), eutrophication potential (EP), freshwater aquatic ecotoxicity potential (FAETP inf.), global warming potential (GWP 100 years), human toxicity potential (HTP inf.), ozone layer depletion potential (ODP), photochemical ozone creation potential (POCP), and terrestrial ecotoxicity potential (TETP inf.). To further analyze the contribution of each impact category to the total environmental burden, normalization, which can provide information about the relative importance of LCIA results [20], was carried out. The normalized factors referred to the ratio of per unit of emission impact divided by the per capita global impact for the year of 2000 [21]. At the same time, weighted evaluation was conducted and the weighted factor was based on the 2012 global life cycle impact assessment survey results of Thinkstep.

3. Results

3.1. LCIA results

The results of LCIA were showed in Table 1 and the squared geometric standard deviation (GSD^2) analyzed by Monte-Carlo method were also presented. The range of each category was between the median multiplied and divided by GSD^2 with a 95% confidence interval, respectively. For the ADP elements category, the environmental impact value was 0.02 kg Sb, GSD^2 was 6.00, indicating that

the value ranges from 0.003 Sb eq. to 0.12 kg Sb eq.. And the value of AP and EP categories were range from 33.09 kg SO_2 to 113.24 kg SO_2 and from 2.74 kg phosphate to 5.52 kg phosphate, respectively. The range of other environmental impact category values can also be calculated by the same method.

The normalized results of HFC-134a production were exhibited in Fig. 2. The HTP category had the most significant impact on the environment. The environmental impacts of ADP fossil, AP, GWP and POCP categories were relatively large, accounted for 44.4% of the overall environmental burden. The remaining impact categories, such as ADP elements and EP, had a smaller impact on the environment, accounting for only 6.1% of the overall environmental burden.

3.2. Main contributors

The contributors of each process to every impact category were showed in Fig. 3. For the categories of ADP element and ODP, chlorine was the main contributor, accounting for 94.6% and 68.7% of the total contributions, respectively. Obviously, except for ADP element and ODP categories, electricity presented a considerable contribution to other impact categories with a share of 36% to 60%. And the contribution of steam to these impact categories ranged from 19% to 33%. In addition, hydrogen fluoride exerted a significant contribution to the categories of AP, FAETP, ODP and POCP, accounting for 32.3%, 28.7%, 25.1% and 21.2%, respectively. The direct emissions contributed 7.4% to the GWP category, and the transportation and other substances contributed very little to all impact categories. Interestingly, the POCP in the transportation stage was negative, which means that driving a truck is beneficial for air quality. This is because that the new version of CML 2001 method in the GaBi software divides the NO_x emissions from trucks into the single emissions NO and NO_2 . NO has a negative effect on the POCP because it decreases the near surface ozone formation.

Fig. 4 showed the relative contribution of significant substances to the five most important impact categories in HFC-134a production by calcium carbide acetylene route. The contribution of coal to ADP fossil category was about 74.9%, of which 46.9% was used for electricity generation, 25.6% for steam production and 13.2% for coke production. For the AP category, the emissions of SO_2 (77.7%) were the main contributor, followed by NO_x (21.0%), both of which were mainly from electricity, steam and hydrogen fluoride. The environmental impact of GWP category was mainly contributed by CO_2 , accounting for 92.7% of the total contribution, while only 6.7% was contributed by CH_4 . The emissions of CO_2 mainly originated from electricity (43.7%), steam (23.9%), chlorine

Table 1
The LCIA results for 1 ton of HFC-134a.

Categories	Value	GSD^2
ADP elements	0.02 kg Sb	6.00
ADP fossil	1.55×10^5 MJ	3.84
AP	61.21 kg SO_2	1.85
EP	3.89 kg Phosphate	1.42
FAETP	65.40 kg DCB	1.39
GWP 100 years	1.29×10^4 kg CO_2	1.74
HTP	3169.13 kg DCB	1.30
ODP	1.04×10^{-7} kg R11	1.19
POCP	4.93 kg Ethene	1.00
TETP	60.25 kg DCB	1.32

Note: GSD^2 is the squared geometric standard deviation.

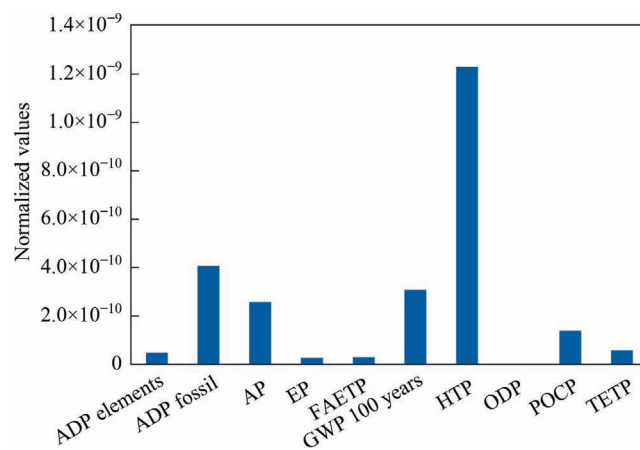


Fig. 2. Normalized results of HFC-134a production.

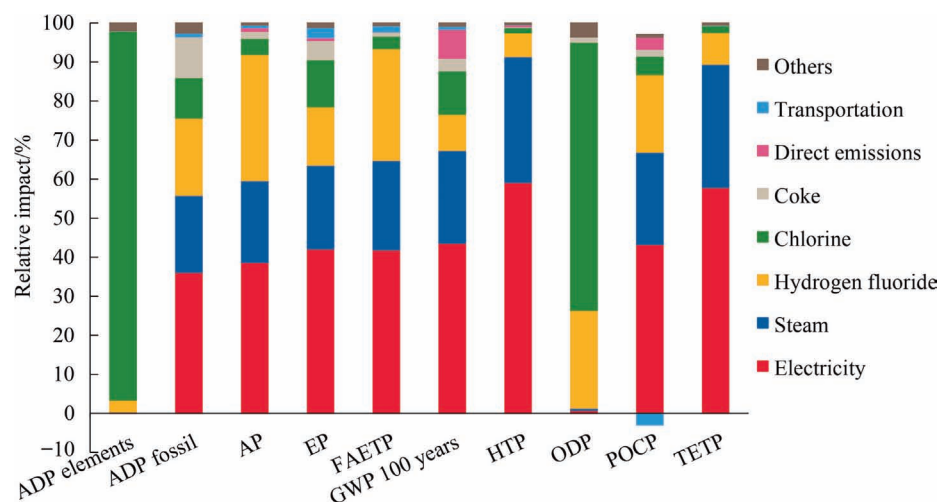


Fig. 3. Relative contribution of each process to every impact category in HFC-134a production.

(11.5%), hydrogen fluoride (9.1%), as well as the direct emissions (8.1%) during the calcium carbide production stage. Pentavalent arsenic (As) and nickel (Ni) discharged into the air contributed the most to the HTP category, accounting for 44.6% and 27.2% of the total contribution, respectively. NMVOC (39.5%), SO₂ (37.7%) and NO_x (12.6%) released in the air had the significant effect on the formation of photochemical ozone. The contribution of the remaining impact categories to overall environmental burden was relatively small, and the corresponding contribution could be obtained from the Supplementary Material S2.

3.3. Sensitivity analysis

Sensitivity analysis can study the variation sensitivity and robustness of the LCIA results and help to identify the key effect factors. Fig. 5 showed the corresponding percentage changes of each environmental impact category when the variation of each input (or each output) parameter was 5%. Sensitivity analysis results indicated that, compared with other elements, coal-based electricity had the most impact on five important impact categories during the entire life cycle. Every 5% decrease in the coal-based electricity consumption reduced the environmental impact of each impact category by 1.8% to 3.0%. As a result, electricity production efficiency significantly reduced the environmental burden of all important impact categories, which was mainly due to the reduction in coal use. The effect of steam was similar to that of electricity. For every 5% decrease in steam use, the environmental impact of each impact category decreased by 1.0% to 1.6%. It is worth noting that the reduction of steam consumption is also mainly due to the reduction in coal use. Every 5% decrease in hydrogen fluoride will bring an environmental benefit of 1.6% in terms of AP category. In addition, with the exception of GWP and POCP categories, the impact of direct emission reductions on other impact categories could be negligible. Therefore, the key to reduce the overall environmental burden of HFC-134a production lies in how to reduce the environmental impact of electricity, steam hydrogen fluoride and chlorine as well as the direct emissions in the production process.

4. Discussion

Electricity had the most significant environmental impact on the entire life cycle of HFC-134a production (Fig. 3). China was one of the largest energy consumers in the world, and its energy

supplies were heavily dependent on fossil fuels. The electricity based on fossil fuels combustion accounted for 69.6% of China's total energy production in 2019 [22]. Coal-based electricity was considered in this study, because of its dominant status in China's energy structure. And the heavy use of fossil fuels had caused serious environmental pollutions in China. Owing to the constraints of fossil fuels and the huge pressure of emission reduction, China must accelerate the development of renewable energy [23]. Renewable energy was expected to gradually replace fossil fuels as the main energy source in China and play an important role in mitigating the impact of climate change and improving energy security [24]. Wind and solar PV power generation have developed rapidly in China since the introduction of the Renewable Energy Act in 2006. They increased from 3.7 TW·h and 0.1 TW·h in 2006 to 405.7 TW·h and 223.8 TW·h in 2019, respectively [25,26], and were expected to increase to 2413.3 TW·h and 2473.0 TW·h by 2035, respectively [27]. Wind and solar energy resources were rich in Northwest China, which were considered to be the most potential alternatives to fossil fuels [28–30]. In addition, wind and solar PV power generation provided considerable benefits in terms of public health and greenhouse gas emissions [31–33]. Therefore, coal-based electricity was replaced by wind and solar PV electricity in this study, and the environmental benefits were very significant, as can be seen from Table 2. In the case of shortage of fossil resources, the ADP fossil category by using wind and solar PV electricity were reduced by 35.6% and 34.3%, respectively. And the GWP category decreased by 43.1% and 41.9%, respectively. For the other important impact categories, the environmental burden would be reduced by 37% to 59%, respectively. The effects of the remaining impact categories were further compared in Supplementary Material S3 in this study. In order to make the research results more intuitive, the ten impact categories were weighted. It can be seen that the overall environmental burden were reduced by 48.4% and 43.6% respectively when using wind and solar PV electricity as substitutes. Accordingly, appropriate adjustment of the power source structure in HFC-134a production could dramatically reduce the environmental burden.

The steam also has a significant impact on the environment during the entire life cycle (Fig. 3). Biomass is a renewable resource, and China is abundant in biomass resources. From 2011 to 2018, the annual output of straw waste increased from 839 million tons to 872 million tons, which is still increasing [34]. The combustion of fossil fuels emitted a large amount of CO₂, while biomass boilers which had broad prospects for development [35,36] were appropriate technologies to reduce greenhouse gas

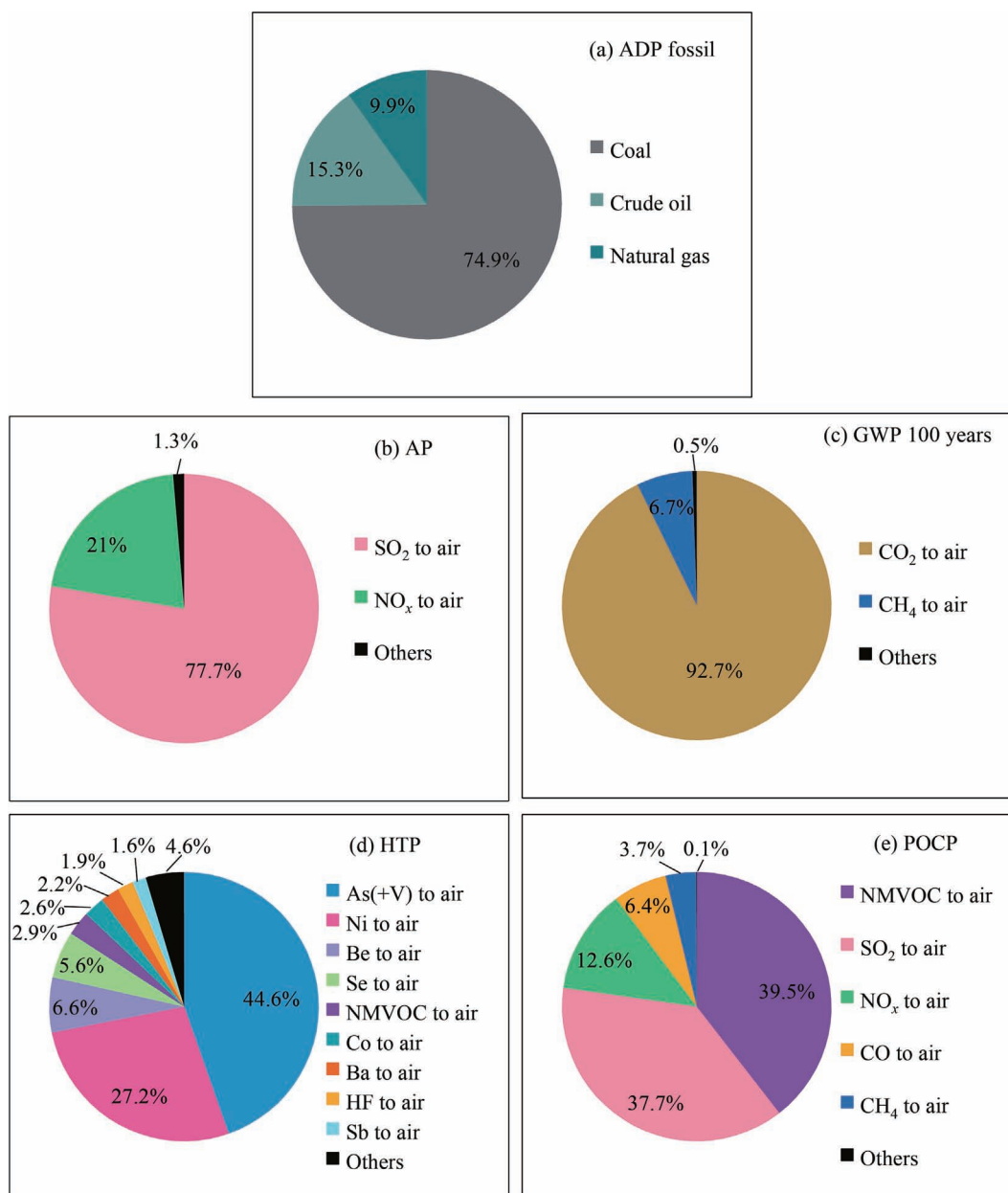


Fig. 4. Relative contribution of significant substances to the five most important impact categories in HFC-134a production.

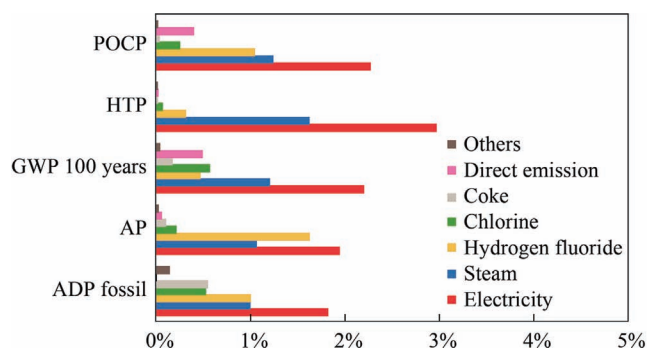


Fig. 5. Sensitivity analysis of HFC-134a production process in change of each input (or each output) parameter.

emissions and increase the proportion of renewable energy [37–39]. Natural gas is a high-quality, efficiency, clean and low-carbon energy, which is usually regarded as a fuel that could be used for energy transformation in order to phase out coal consumption in cogeneration plants [40,41]. So in this study, biomass and natural gas were used instead of coal to produce steam. As shown in Table 2, the use of biomass steam had significant environmental benefits to the categories of ADP fossil, GWP and HTP, reducing the environmental burden by 18.7%, 22.8% and 24.8% respectively, compared to coal-fired steam. Although the use of natural gas boilers for steam production increased fossil fuel consumption increased by 2.2%, for other important impact categories, the environmental burden were all reduced to varying degrees. In particular, the environmental impact of HTP category reduced by 31.3%. According to the weighted results, the total environmental burden of biomass steam and natural gas steam was about 20% less than that of coal-fired steam. Although the environmental benefits

Table 2

Comparison of LCIA results when using different types of electricity and steam in HFC-134a production.

Categories	Coal-based energy	Wind electricity		Solar PV electricity		Biomass steam		Natural gas steam	
		Value	Variation	Value	Variation	Value	Variation	Value	Variation
ADP fossil/MJ	1.55×10^5	9.95×10^4	–35.6%	1.02×10^5	–34.3%	1.26×10^5	–18.7%	1.58×10^5	+2.2%
AP/kg SO ₂	61.21	37.75	–38.3%	38.34	–37.4%	55.63	–9.1%	50.1	–18.3%
GWP 100 years/kg CO ₂	1.29×10^4	7338.19	–43.1%	7490.52	–41.9%	9964.02	–22.8%	1.19×10^4	–8.0%
HTP/kg DCB	3169.13	1321.35	–58.3%	1487.52	–53.0%	2384.71	–24.8%	2178.58	–31.3%
POCP/kg Ethene	4.93	2.67	–45.8%	2.78	–43.7%	4.81	–2.6%	4.04	–18.2%
Weighted results	1.78×10^{-8}	9.21×10^{-9}	–48.4%	1.01×10^{-8}	–43.6%	1.43×10^{-8}	–20.1%	1.43×10^{-8}	–19.6%

Note: “+” and “–” respectively represents the increased and decreased environmental burden compared to the coal-based energy (coal-based electricity and coal-fired steam).

brought by the two were comparable, China was short of natural gas resources and its external dependency reached 45.3% in 2019 [42]. Moreover, China was abundant in biomass resources, but approximately 65% of straw was burnt or discarded every year, which not only brought huge waste of resources, but also caused serious environmental pollution [43]. Biomass boilers, with obvious advantages in reducing fossil energy use and emissions of CO₂, SO₂, and NO_x [44–47], are more in line with China's current energy strategy requirements and environmental protection requirements.

The production of hydrogen fluoride and chlorine has a relatively large impact on the environment. The production methods of chlorine mainly include ion membrane method, mercury method and diaphragm method. China has basically eliminated the backward mercury method and diaphragm method. In this study, the more advanced ion membrane method was used to produce chlorine. The data of three chlorine production methods were all obtained from the GaBi database. It can be seen from Fig. 6, the environmental benefits of the ion membrane method were better than the other two methods. Therefore, the application of advanced production technology can reduce the impact on the environment. China basically uses the fluorite method to produce hydrogen fluoride, which has high energy consumption. In order to reduce its impact on the environment, in addition to improving its own production technology, the potential of reducing the raw materials consumption should also be actively explored. In particular, the actual use of hydrogen fluoride in the HFC-134a production stage is about 7% more than the theoretical requirement.

Direct emissions from HFC-134a production contributed 7.4% to the GWP category (Fig. 3). The main reason is that the lime tail gas in the calcium carbide production stage contains a large amount of CO₂, which is directly discharged into the air, accounting for 8.2% of CO₂ emissions in the entire life cycle. And this part of CO₂ is expected to be reduced through carbon capture, utilization and

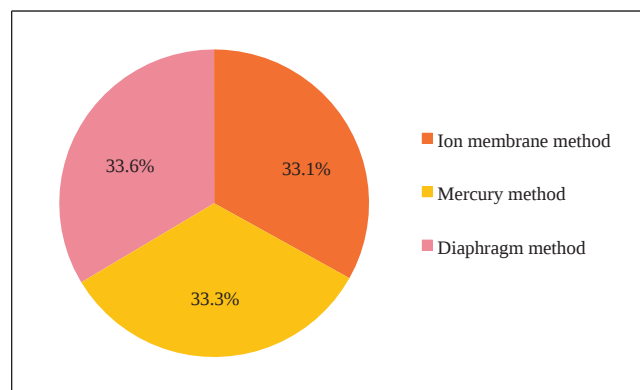


Fig. 6. Comparison of weighted results when using different methods to produce chlorine in HFC-134a production.

Table 3Change of LCIA results after using CO₂ capture and purification technology in HFC-134a production.

Categories	Variation value	Variation
ADP fossil/MJ	+139.00	+0.1%
AP/kg SO ₂	+0.11	+0.2%
GWP 100 years/kg CO ₂	–527.10	–4.1%
HTP/kg DCB	+3.87	+0.1%
POCP/kg Ethene	+0.01	+0.2%

Note: “+” and “–” respectively represent the increased and decreased environmental burden after using CO₂ capture and purification technology to capture CO₂ in lime tail gas.

storage (CCUS) technology which can reduce CO₂ emissions into the atmosphere [48,49]. In 2019, China has 18 CO₂ capture projects in operation, with CO₂ capture capacity of approximately 1.7 million tons, as shown in Supplementary Material S4 [50]. This technology is not only used in coal-fired power plants, but also in industries such as coal-to-liquid, coal-to-methanol, and fertilizer production. In addition to geological storage, most of the captured CO₂ is used to enhanced oil recovery (EOR) and sell after refining, which can not only reduce CO₂ emissions, but also produce economic benefits. As the world's largest CO₂ emitter [51], the successful operation of these projects is of great significance to promote China's carbon emission reduction, and it is also a great inspiration to the application of CCUS technology in the calcium carbide production industry. In this study, on the basis of the case system, the CO₂ capture and purification technology was used to capture CO₂ in lime tail gas to obtain CO₂ products (see Supplementary Material S5 for the life cycle inventory). Although new materials and energy had been introduced, for every ton of HFC-134a produced, the CO₂ equivalent emissions were reduced by 527.10 kg, the value of GWP category reduced by 4.1%, while the environmental impact of other important impact categories only increased by 0.2% (Table 3). Therefore, when using calcium carbide acetylene method to produce HFC-134a, applying the CO₂ capture and purification technology to capture CO₂ in lime tail gas performs well in terms of environmental benefits, which is in line with China's low-carbon development strategy.

5. Conclusions

This study quantified the environmental performance of HFC-134a production by carrying out a life cycle assessment using the CML 2001 method, identified the key factors affecting the environment, and proposed ways to improve the environmental benefits. And the sensitivity and uncertainty assessments were also implemented to improve the credibility of the current work. The results were as follows:

The overall environmental burden of HFC-134a production were mainly contributed by five impact categories (HTP, ADP fossil,

GWP, AP and POCP). Electricity and steam had significant impacts on the environment. The environmental impacts of hydrogen fluoride and chlorine were relatively large. And direct CO₂ emissions in the calcium carbide production stage accounted for 8.2% of CO₂ emissions in the entire life cycle.

The overall environmental burden of HFC-134a production can be reduced by replacing the use of coal and adopting advanced production technologies. The overall environmental burden were reduced by 48.4% and 43.6% respectively when the wind electricity and the solar PV electricity were used instead of coal electricity. And the overall environmental burden were reduced by about 20% when biomass steam and natural gas steam were used instead of coal-fired steam. Compared with mercury method and diaphragm method, the overall environmental burden of HFC-134a production were reduced by 0.2% and 0.5% by using more advanced ion membrane method for chlorine production.

Moreover, using CO₂ capture and purification technology to capture CO₂ in lime tail gas performs well in terms of environmental benefits. At this point, the environmental impact of the GWP category would be reduced by 4.1%, while the environmental impact of other important impact categories would increase by only 0.2%.

HFC-134a can be not only used as a refrigerant, but also used in medicine, pesticide, and cleaning industries. Studying the environmental performance of HFC-134a production by LCA method is very important for the current development of the industry. This study can expand the LCI database of HFC-134a production, provide useful information for producers, and offer a reference for decision-making to promote the sustainable development of the HFC-134a industry.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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