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Environmental impact assessment of generating cold atmospheric pressure plasma and plasma-activated water at lab scale

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Cold atmospheric pressure plasma (CAPP) is the fourth state of matter produced by applying high energy to gas, and water treated with CAPP is known as plasma-activated water (PAW). CAPP and PAW have shown successful applications in food safety and functional modifications. These novel technologies are not commercially applied in the food industry yet as their sustainability benefits are not fully understood. This study assessed the carbon footprint of producing CAPP and PAW on a lab scale. CAPP produced 7.9×10^{-3} kg CO_{2e} per 1 min of plasma generation time, while PAW produced 7.9×10^{-2} kg CO_{2e} per 10 min of plasma generation time, with the majority of greenhouse gases (GHGs) being generated by electricity sources. Adopting the wind or solar energy as a renewable electricity source could substantially reduce the carbon footprint of CAPP and PAW. This study provides valuable insights to guide the future commercialization of cold plasma as a sustainable food preservation technology.

Introduction

Plasma is the fourth state of matter and consists of ions, electrons, and uncharged particles such as atoms, molecules, and radicals present in a partially ionized gas form radicals.¹ Traditionally, cold plasma processes were used for coating, microelectronics, machinery, optics, *etc.* and were conducted under vacuum, which is not an energy-efficient method for the food industry.² In recent years, non-thermal plasma or cold atmospheric pressure plasma (CAPP) discharges can be produced at ~1 atm under ambient conditions, thus simplifying and reducing the cost of industrial applications without the need to maintain the pressure.³ In the food industry, CAPP treatment can be applied for microbial inactivation of food and food contact surfaces (packaging and equipment), for functional and structural modifications (changes in the texture and nutritional yield), as well as for environmental disinfection.^{4–8} An indirect mode of using CAPP is to treat liquids, such as water, in plasma, which leads to the transfer of reactive species

Sustainability spotlight

As a novel non-thermal food technology, there is a need to understand the environmental impacts of plasma, which has not been investigated in the food sector so far. The carbon footprint of lab-scale production of cold atmospheric pressure plasma (CAPP) and plasma-activated water (PAW) was quantified in this study. The majority of greenhouse gases were generated from the existing electricity grid, which can be substantially reduced if transitioned to renewable sources. Our results provide a starting point for non-thermal plasma researchers and food industries looking for sustainable sanitization technologies, aligning with UN's Sustainable Development Goal #12 – Responsible Consumption and Production.

from plasma to the water medium, thus changing the chemistry and properties of water.⁹ This water is termed as plasma-activated water (PAW). Fresh produce need to be washed after harvesting to remove dirt and reduce the microbial load. In this case, a direct application of CAPP may not be effective and feasible due to the absence of a liquid medium. PAW can overcome this challenge and can potentially replace the chemical liquid sanitizers used in the food industry. Both CAPP and PAW are heavily researched in food applications and have successfully shown microbial inactivation on various food and food contact surfaces.^{3,10–16} PAW can be prepared on demand and on-site, and it does not need to be diluted unlike conventional sanitizers such as chlorine and quaternary ammonia, which require special transportation, storage, handling, and dilution before application. CAPP has the same advantages as PAW, and in addition, it eliminates the need for water. This reduces water usage in food sanitation as well as potential wastewater treatments that are associated with conventional liquid sanitizers.

The rapidly growing global population increases the demand for food, water, and energy resources. Hence, sustainability and long-term ecosafety are being investigated for existing and novel technologies used in food and agriculture.¹⁷ Food production accounts for approximately one-third of global anthropogenic greenhouse gas emissions.^{18,19} This has led to a variety of research efforts focusing on the development of healthy,

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quality, and safe food products with more environmentally friendly processes.²⁰ Life cycle assessment of novel food preservation technologies such as high hydrostatic pressure, microwave, modified atmospheric packaging, and osmotic dehydration has been reported in the literature.^{21,22} As mentioned previously, CAPP and PAW are widely researched for various applications on lab scales, but the next stage of research requires scale-up, dosage requirements, cost analysis, and life cycle assessment (LCA) to evaluate their broader impacts. To our knowledge, there is only one LCA study using plasma for face mask decontamination,²³ and no LCA studies have been conducted for plasma applications in the food sector. Although there are some scale-up studies coming up for plasma applications, it is generally assumed that plasma technology is green and sustainable.²⁴ Hence, it would be beneficial to the industry to understand the sustainability aspects before adopting this technology. The objective of this study is to assess the carbon footprint of CAPP and PAW generated on a lab scale. We aim to numerically understand the environmental impact of this novel non-thermal technology, which has not been investigated yet.

Materials and methods

Goal and scope

This study is intended for researchers exploring the applications of novel non-thermal processing technologies like plasma. Given the primary focus of this study is to evaluate the impacts of CAPP and PAW production, a gate-to-gate system boundary was selected, starting from plasma generation to CAPP or PAW production. This study excludes the downstream application of these processes. The functional unit (FU) of this study is the plasma generation time in minutes. We used different FUs for CAPP and PAW based on required microbial reduction. For CAPP, this FU (1 min) is the time required for microbial inactivation of organisms when food or surfaces are directly treated with plasma; in terms of PAW, this FU (10 min) is the minimum time required to treat water with plasma to have a desired concentration of reactive species and acidity of PAW to make it suitable for microbial inactivation applications. Since this study focuses on the lab-scale generation of CAPP and PAW using one specific plasma equipment, we acknowledge that the findings

from this study may not be generalized to all plasma equipment since different plasma equipment vary in terms of design, operational conditions, and performance characteristics. Each equipment's unique configurations can lead to different interactions with water and food products, affecting the overall microbial inactivation performance. Given the goal of this study is to determine the carbon footprint of the production of lab-scale CAPP and PAW in Raleigh, North Carolina, we have adopted the Southeastern Electric Reliability Council (SERC) Virginia/Carolina electricity mix as the main electricity source. A sensitivity analysis was also performed by exploring renewable energy sources to evaluate the potential environmental impacts of switching to a clean electricity grid. In North Carolina, two policies related to renewable energy sources: Solar for All Program (Energize NC) and Executive Order No. 218 by the NC Governor, highlight the encouragement of installation of solar and wind power, respectively.^{25,26} Hence, these two renewable energy sources were chosen as potential future clean energy sources to generate CAPP and PAW as a part of sensitivity analysis.

Life-cycle inventory and impact assessment

In this study, primary data were used as the major source for compiling the inventory database. Table 1 shows major input and output values calculated for CAPP and PAW based on their specific generation times. Specifically, all the input values were provided by the equipment supplier (Plasmatreteat, USA) in the USA, and the emission data were obtained from the equipment supplier (Plasmatreteat GmbH) in Germany. Plasma was generated using an Openair® plasma system consisting of an RD1004 jet and FG5001 plasma generator from Plasmatreteat, USA, Inc. Dry, filtered, and compressed air at room temperature (average flow rate of 33.5 L min⁻¹) was used as the feed gas with the power consumption of 465 W. The fluctuation in power supply is not addressed in this study as these are the typical values provided by the equipment manufacturer. Since we only have 2 input parameters (air and electricity), a sensitivity analysis was only performed on electricity, assuming air would not show major fluctuations. The high voltage triggered the release of plasma at high velocity through the rotating nozzle (#22892 > 2000 rpm). The 1 min plasma generation time was selected

Table 1 Major inputs and outputs of CAPP and PAW generation at lab scale

	CAPP (FU: 1 min generation time)	PAW (FU: 10 min generation time)	Units
Input			
Power	7.75×10^{-3}	7.75×10^{-2}	kW h
Air flow	33.5	335	L
Deionized water (DI)	N/A	200	mL
Output			
Plasma generated	1	N/A	Min
Plasma-activated water generated	N/A	190	mL
NO _x	0.3166	3.166	g
O ₃	3.33×10^{-5}	3.33×10^{-4}	g
H ₂ O vapor	N/A	10	mL



based on 3.9 log reduction achieved for adherent *Escherichia coli* cells on stainless steel by CAPP treatment when this equipment was used in our lab.⁸ In the case of PAW production, a 10 min production time and 200 mL water were used based on >5 log reduction of *Salmonella typhimurium* on shell eggs achieved after using the water that was treated with plasma for 10 min by a study conducted in our lab.¹⁵ This PAW had a pH of 2.80 and a nitrate concentration of 10.35 mM (641.8 ppm). The same study also showed that a lower PAW generation time led to lower microbial inactivation in planktonic cells of *S.typhimurium*. Other studies that used the same equipment to generate PAW also showed a 5 log reduction of *Salmonella typhimurium* and *Klebsiella michiganensis* for 10 min and 5 min generation times, respectively.^{27,28} In food or food-contact surface sanitization, a 3–5 log reduction is considered a desirable log kill. Hence, 10 min generation time for PAW was selected as the best

available data in this study. Deionized water (DI) was used in this study as it has been widely adopted for making PAW in the published literature.²⁹ The studies conducted by our lab as well as by the plasma equipment manufacturer showed a 5–10% water loss due to evaporation when generating PAW, attributed to the temperature increase to 50–60 °C. Hence, 10 mL of water vapor was considered a loss to the air. The energy consumption data were converted to kWh based on CAPP and PAW generation times. We assume that the plasma generation takes place under a stable power supply and that equipment operates efficiently and properly. As for the emissions, nitrogen oxides (NO_x) and ozone (O₃) were the only gases detected during plasma generation processes, while other emissions were assumed to be negligible. NO_x and O₃ were measured directly using PG-250 and APOA-360, respectively (HORIBA Europe GmbH, Oberursel), at Plasmatrete in Germany. In this study, few inputs with a small variation of inventory data (<5%) were reported by the manufacturer; given the limited variability, no other uncertainty analysis such as Monte Carlo simulation was performed.

In addition to primary data, ecoinvent v3.8 was used to cover the background data gaps such as electricity sources for Virginia/Carolina (Southeastern Electricity Reliable Council or SERC). Once we compiled the inventory data, OpenLCA 2.0.3 software was used in this study to develop LCA models. TRACI 2.1 (Tool for the Reduction and Assessment of Chemical and other environmental Impacts) is an environmental impact assessment method developed by the U.S. Environmental Protection Agency. TRACI 2.1, utilizing IPCC AR4 on a 100-year time horizon, was selected to calculate the global warming potential (GWP) in kg CO_{2e} per FU.

Results and discussion

Overall, as seen in Fig. 1, CAPP production has less carbon footprint of 7.9×10^{-3} kg CO_{2e} per FU than PAW production (7.9×10^{-2} kg CO_{2e} per FU), largely due to its lower generation time required to achieve microbial reduction. In addition, CAPP production does not involve water usage, residual waste, or the need for waste treatment, further reducing its environmental footprint. Table 2 outlines the breakdown of GWP results of CAPP and PAW when using the SERC as the base case electricity source. We found that the major contribution (~52%) for the carbon footprint came from electricity consumption, followed

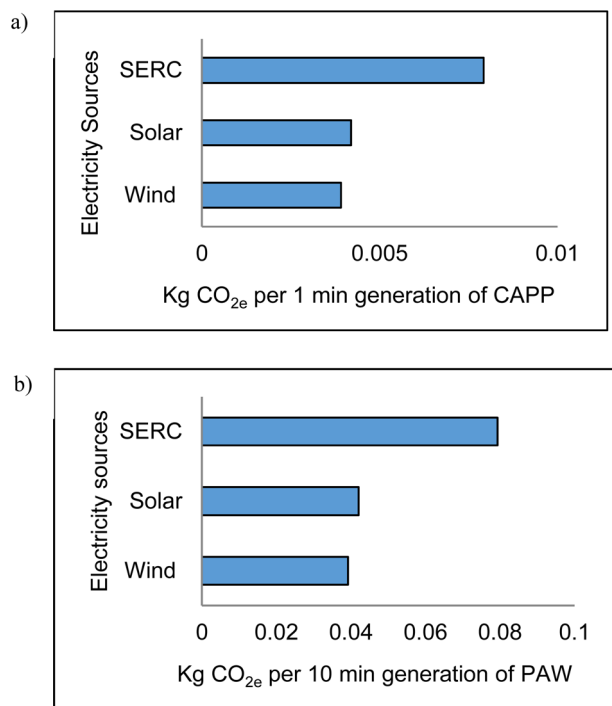


Fig. 1 Environmental impact assessment results of (a) CAPP and (b) PAW generation from different electrical sources.

Table 2 Percent contribution to kg CO_{2e} in generation of CAPP and PAW

Process	Total kg CO _{2e}
CAPP (FU: 1 min plasma generation time)	
Market for electricity, high voltage electricity, high voltage cutoff, U – US-SERC	4.1×10^{-3} (51.8%)
Market for compressed air, 600 kPa gauge compressed air, 600 kPa gauge cutoff, U – RoW	3.8×10^{-3} (48.2%)
Total	7.9×10^{-3}
PAW (FU: 10 min plasma generation time)	
Market for electricity, high voltage electricity, high voltage cutoff, U – US-SERC	4.11×10^{-2} (51.8%)
Market for compressed air, 600 kPa gauge compressed air, 600 kPa gauge cutoff, U – RoW	3.82×10^{-2} (48.1%)
Market for water, deionised water, deionised cutoff, U – RoW	1×10^{-4} (0.11%)
Total	7.9×10^{-2}



by compressed air (~48%) for both CAPP and PAW. Coal (~19%) and natural gas (~16%) were major contributors to carbon footprint when using SERC as the electricity source. Sensitivity analysis on electricity sources further showed that when SERC was replaced with 100% wind or solar energy, the total carbon footprint could be reduced significantly to 4.0×10^{-3} kg CO_{2e} and 4.0×10^{-2} kg CO_{2e} per FU of CAPP and PAW generation, respectively (Fig. 1). Solar energy also included natural gas as one of the contributors, but wind energy did not have natural gas or coal. Hence, using alternative energy sources in producing CAPP and PAW can be beneficial in reducing environmental impacts. It is also worth noting that with the development of technologies, compressed air becomes less necessary for some plasma devices in various labs, including ours. Thus, in the future, there is a potential to drastically reduce the environmental impacts by eliminating compressed air and using alternative electricity sources. This study utilized two generation times for functional units because when you use CAPP directly on food or food contact surfaces the time required is lower compared to using it indirectly (in the form of PAW) for required microbial reduction. This provides a comparison to readers when using CAPP and PAW for food sanitizing applications. Unlike other technologies, there is no dosimetry for CAPP and PAW, and different equipment may result in different concentrations of CAPP and PAW. Hence, a microbial reduction of 3–5 log was used to standardize the CAPP and PAW treatments.

The carbon footprint results from our study are based on one specific equipment that uses a gliding arc mechanism and compressed air for food sanitizing applications. The environmental sustainability of CAPP and PAW generation can be improved by optimizing these processes to suit their specific applications. For instance, the generation times of CAPP and PAW can be modified based on application requirements and by changing electrical parameters. Adding microbubble generation during PAW production enhances the mass transfer of reactive species from the gas to liquid.³⁰ This can improve the efficacy of PAW and potentially reduce the generation time of 10 min in the future depending on the application of PAW. The generation time of PAW can also be reduced if a lower pH (<3) is not desired. Additionally, different plasma equipment may be needed for various applications, and each design can have distinct power consumption requirements. By optimizing the characteristics of plasma equipment for each application during scale-up, the treatment time and emissions can be potentially reduced as well.

Compared to chlorine, the most common sanitizer used in the food industry, using CAPP directly on a food product or equipment offers a streamlined approach to sanitation. It can effectively eliminate the complicated procedures associated with chlorine, including special handling, transport, dilution, and an additional step of activated carbon to remove chlorine residues before being disposed of into sewage. CAPP can be prepared on-site and on-demand, and there is no waste generated; thus, no special disposal is needed. Similar to CAPP, PAW can also be prepared on-site and does not require any special handling or dilution before use. The wastewater from PAW can

either be neutralized with a base (such as caustic) or flushed with water, as our nitrate concentrations are extremely low (~0.07%), as indicated in this study.³¹ Therefore, although gases like NO_x and O₃ are associated with plasma generation, their environmental impacts can be minimized by either utilizing gas scrubbing systems or catalysts to break them down into less harmful components. In contrast, conventional sanitization methods like chlorine may present another environmental challenge such as the treatment of chlorine-containing wastewater.

So far, there are no published studies yet to understand the environmental impacts of plasma and PAW generation for food applications. The only plasma-related LCA study was conducted by Sinkko *et al.* (2023) in medical applications.²³ They reported that single-use face masks had a lower environmental impact than using a decontamination process with plasma due to the use of helium for the plasma generation process. Other LCA studies using emerging technologies in food processing typically adopt a broader system boundary including downstream application steps. For instance, Pardo *et al.* (2012) conducted LCA on all steps involved in food processing (cooking and packaging).²² They reported reduced environmental impacts in terms of energy demand and CO₂ emissions by emerging technologies (high pressure processing and microwave) compared to conventional pasteurization. Lower water requirements were also noted by high pressure processing when compared to thermal processes.²² Given the early-stage of plasma-related LCA studies in food processing, it will be beneficial to broaden the scope of our study in the future. The findings from this LCA study may not be generalized to all plasma equipment and large-scale generation of CAPP and PAW since this study focuses on the lab-scale generation of CAPP and PAW using one specific plasma equipment. Nevertheless, this study provides a starting point for plasma researchers aiming for scale-up applications with key considerations to prioritize resources and efforts. An optimized commercial process may end up using resources such as electricity more efficiently compared to lab scale, resulting in less carbon footprint per sanitization treatment. Future LCA study can expand the scope of our work to include an optimized process model and the application steps of CAPP and PAW in food/food contact sanitization to provide a more comprehensive understanding of the sustainability of plasma-based technology in the food sector.

Conclusions

In summary, we have assessed the carbon footprint of generating lab-based CAPP and PAW for food and food contact sanitization applications. The environmental impact in our case was mainly from the electricity source, which can be reduced substantially when transitioning to renewable electrical sources. Our results provide a starting point for non-thermal plasma researchers and food industry looking for sustainable sanitization technologies. Further investigation of CAPP and PAW with other non-thermal technologies and with existing industry sanitizers is essential to ensure the wide adoption of plasma-based technologies in food systems.



Data availability

This study was carried out using the publicly available database from ecoinvent v3.8 with OpenLCA 2.0.3 software.

Author contributions

Urvi Shah: conceptualization, data curation, formal analysis, methodology, writing – original draft & editing. Minliang Yang: conceptualization, formal analysis, software, supervision, writing – review & editing. Deepti Salvi: conceptualization, funding acquisition, supervision, project administration, writing – review & editing.

Conflicts of interest

There are no conflicts to declare.

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